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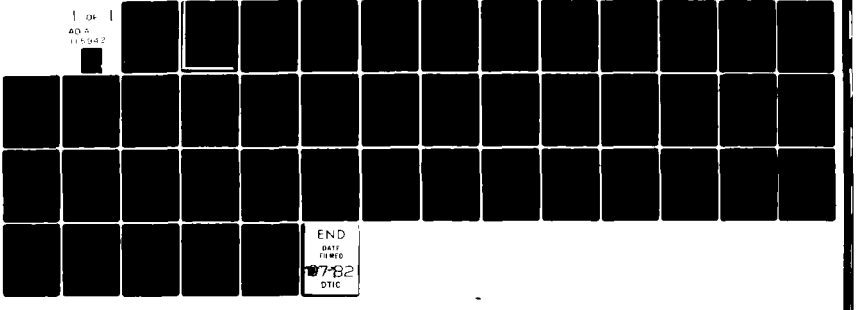

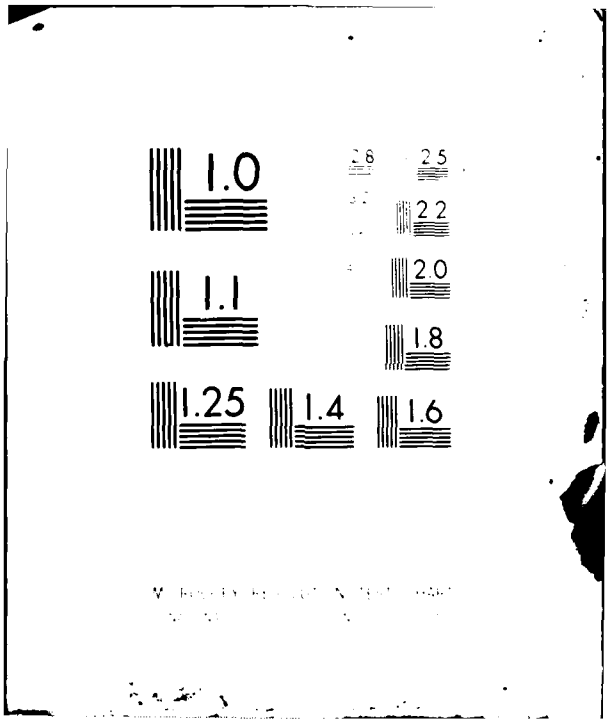
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FINAL REPORT

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Aspects of High-Energy Laser Theory

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Prepared by: Mark Hillery

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The research which was conducted under this contract fell mainly into three categories: gain switching, infection locking, and photon statistics. In addition, some work in the foundations of radiative transport theory and spectroscopy was also performed. The use of a metastable state as the upper laser level was shown to offer the possibility of obraining large inversions and thereby storing large amounts of energy in the lasing medium. The problem associated with this is that the linear gain which one obtains from the transition from this level to the lower laser level is quite small. The possibility of "dumping" the energy stored in the metastable was investigated.

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The research which was conducted under this contract fell mainly into three categories: gain switching, injection locking, and photon statistics. In addition, some work in the foundations of radiative transport theory and spectroscopy was also performed. A brief summary of this work will now be given.

I. Gain Switching

The use of a metastable state as the upper laser level offers the possibility of obtaining large inversions and thereby storing large amounts of energy in the lasing medium. The problem associated with this is that the linear gain which one obtains from the transition from this level to the lower laser level is quite small. The possibility of "dumping" the energy stored in the metastable level by the application of an external field was investigated.[1] The external field, by inducing a dipole moment, increases the transition probability between the two levels. One can, therefore, obtain a large inversion and then switch on the field to obtain a good value for the linear gain.

An estimate of M1, E2, and induced E1 transitions (for an applied electric field of 10^5 Vcm^{-1}) indicated that transitions satisfying the following two requirements would be good candidates for gain switching:

A) they are forbidden for M1 radiation

B) they occur at wavelengths of 1 to 10 μm

Vibrational transitions in homonuclear diatomic molecules satisfy these conditions. For H_2 one can obtain a gain coefficient, K, of

$$K = \frac{\Delta N}{N} (5 \times 10^{-2}) \text{ cm}^{-1}$$

where $\Delta N/N$ is the relative population inversion and the electric field is assumed to be 10^5 V/cm.

If one tries to use electric fields much larger than those considered above, breakdown will occur. For the alternating electric field produced by a giant laser pulse it is possible to obtain much higher values of the field. This would allow the gain to be increased by two orders of magnitude.

Spin forbidden transitions were also examined to see if they would be of use in gain switching schemes. In particular the transition from the $^1\Delta$ state of molecular oxygen to the ground state was considered because this state can be produced in chemical reactions so that a practical pumping scheme is available. An inhomogeneous magnetic field will cause spin multiplet intercombination and, therefore, make spin forbidden transitions possible. Actual calculations for the $^1\Delta$ - oxygen system, however, showed that the necessary magnetic fields were not practically obtainable.

II. Injection Locking

There are applications where it is necessary to have a high energy laser with a particular polarization. Due to practical considerations it is not always possible to use intracavity polarization selective elements. The laser output is then unpolarized, or because of resonator geometry or reflective properties of the intracavity optical surfaces, the laser may have a prefered polarization which is usually not known apriori, and in some cases, is time varying.

It is possible that injection locking, which has been successfully applied to frequency control lasers, may also be used for polarization control. In fact, experiments have been performed where a polarized output was obtained by injection locking an unpolarized rare gas halide laser. We wanted to determine if polarization selection by injection locki will work for a wider range of lasers.

We have extended the semiclassical laser theory to include laser operation with different polarizations and with an injected signal. We considered two problems. The first is the effect of a polarized injected signal on an unpolarized laser. The second is the effect of a polarized injected signal on a laser that has a prefered polarization. We wanted to know if the injected signal can force the laser into operating with a different polarization. This latter case will be of interest in the scaling of lasers by using a multiplex array. It may also find applications in lasers with annular gain regions which at present operate with axicon type resonators that produce outputs that have time varying polarizations. Our goal was to obtain as understanding of the mechanisms underlying polarization control via injection locking. As such, we have not dealt with the more computationally complicated though possibly more useful cases of operation in the strong-signal regime and with an unstable resonator. We are, at present, investigating the above two problems.

Our results indicate that, except when the injected signal and free running laser polarizations are orthogonal to each other, it is not possible to rotate the laser polarization so that it lines up with the injected signal polarization. However, the difference in polarization may be made arbitrarily small by increasing the injected signal strength. The reason for $\theta \neq \theta_i$ for finite injected signal strength and $0<\theta_i<\pi/2$ may be seen by resolving the injected signal electric field into a component that is parallel to the free running laser polarization and component that is perpendicular to it. The parallel component has a greater effect per electric field strength than the perpendicular component because of the higher laser gain in that direction. Consequently, the final laser polarization will always be between the free running laser and the injected signal polarizations. When $\theta_i = \pi/2$, there is no parallel component of the injected original and therefore it is possible to make $\theta = \theta_i$.

If we consider the situation where we have an array of lasers and where each laser has a different preferred polarization (i.e. the output of the free running laser array is unpolarized), then the distribution of polarization directions of the phase locked array will have a width given by

$$\Delta\theta \simeq \frac{1}{2}\frac{\Delta\alpha}{\Delta c}\sqrt{\frac{I}{\zeta}}$$

where $\Delta\alpha$ is the difference in the gain for the components of the polarization parallel and perpendicular to the injected polarization, $\Delta c = c/2L$ where L is the length of the laser, I is the intensity of the laser inside the cavity, and ζ is the intensity of the injected signal outside the cavity.

III. Photon Statistics

Our group has had an interest in photon statistics problems for several years. Recent work includes a generalization of previous results for a single mode m-photon absorbtion process to an arbitrary number of modes.[2] Two-photon lasers were also studied. The photon statistics of such a laser with a loss mechanism simulated by two-level atoms which absorb a single photon was found.[3] This was then used to investigate nonclassical effects in the light emitted.[4] Two-mode lasers were investigated as well and the intensity correlations for a laser of this type with coupled transitions were found.[5]

IV. Other Work

Fundamental work in spectroscopy was also done. It was shown that time-delayed measurements allow one to obtain accuracy greater than the natural linewidth in the measurement of spectral lines.[6] The foundations of the theory of radiative energy transfer were also the object of a study.[7]

Wave propagation in a random medium was considered and various quantities appearing in the usual phenomenological theory were related to the stochastic properties of the medium.

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GAIN SWITCHING IN HIGH POWER LASERS *

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We consider situations whereby energy could be stored in a metastable state and then "dumped" by switching on an external field thus enhancing the coupling to a lower state at some later time. The present calculations indicate that this concept merits experimental study.

In many high power lasers, one often seeks a population inversion between levels which are only weakly coupled in order that a substantial inversion may be obtained without excessive decay to the groundstate via spontaneous emission. In some cases, however, this leads to the complication that the linear gain associated with the transition between these two levels is very small. It is thus interesting to consider situations whereby energy could be stored in a metastable state and then "dumped" by switching on or enhancing the coupling to a lower state at some later time. This might be accomplished, for example, by applying a strong electric field to a metastable state. This "field induced" effect is well understood theoretically [1,2], and has been verified in numerous experiments [5,6,14,16]. Thus we propose to use field induced transitions as a technique for switching the gain in an inverted medium by the application of an external field. This technique could also be applied to amplifier systems. Furthermore, it would allow one to precisely control the onset of lasing action.

In this paper we shall demonstrate the feasibility of this concept by studying a specific type of molecular transition, and show that gains of several percent per cm can be obtained. The present calculations indicate that the concept of gain switching deserves further experimental study.

In the present calculation we wish to compare the electric dipole transitions which are induced via an external field (induced dipole IE1) with other types of allowed transitions, leading to magnetic dipole (M1) and electric quadrupole (E2) radiation. The orders of magnitude involved in these processes are well known in the M1 and E1 cases and can be easily estimated in the IE1 case. For IE1 transitions the dipole moment induced by external field F is given by [3]:

$$d_F = \alpha F,$$

$$\alpha = \sum_r \langle b|D|r\rangle\langle r|D|a\rangle\left(\frac{1}{E_r - E_b} + \frac{1}{E_r - E_a}\right) = \langle b|\alpha|a\rangle, \tag{1}$$

where D is the electric dipole operator; a, b and r label the lasing and intermediate states in the transition considered. Tensor α is the matrix element of some scattering tensor operator α [4]. Actually formula (1) gives the symmetric part of α only, which can be shown to dominate the antisymmetric part in most cases. The

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scattering tensor may be estimated in terms of an average transition energy ΔE and typical dipole matrix element d as follows

$$\alpha \simeq 2\frac{\langle b|D\cdot D|a\rangle}{\Delta E}. \qquad (2)$$

$$d_F \simeq 2\frac{\langle b|D\cdot D|a\rangle}{\Delta E}F \simeq 2\frac{d^2}{\Delta E}F \simeq 4\times 10^{-4}\,d. \qquad (3)$$

Here we have assumed an external electric field $F = 10^5$ V cm^{-1}, a typical transition energy $\Delta E \simeq 4 \times 10^{-12}$ erg corresponding to radiation at optical wavelength $\lambda = 0.5$ μm and a dipole matrix element of the order of magnitude $d \sim ea_0$. Estimates of this type lead to rigorous bounds on the scattering tensor in all cases where the transition matrix element $\langle b|\alpha|a\rangle$ can be calculated in terms of diagonal matrix elements [4]. This occurs for example for atomic transitions between fine splitting components in LS-coupling and for vibrational transitions in molecules. α may be appreciably larger than estimated in (3) if there are strongly resonant levels r close to any of the levels a or b. In general however (3) gives the correct order of magnitude for atomic systems.

For M1 transitions the magnetic dipole moment is a multiple of the Bohr magneton [3]:

$$d_{M1} \simeq eh/2mc = \tfrac{1}{2}(e^2/hc)ea_0 \sim 3.6\times 10^{-3}\,d. \qquad (4)$$

Finally for E2 transitions the rate is obtained as follows [3]: Neglecting numerical factors of order unity we replace the dipole matrix element d by the electric quadrupole moment Q and divide by the wavelength λ of the radiation emitted. Since Q equals roughly a typical intra-atomic distance times the dipole moment we obtain for the corresponding matrix element:

$$d_{E2} = \lambda^{-1}Q = \lambda^{-1}a_0 d \sim 10^{-4}\,d \quad \text{for } \lambda = 0.5\ \mu\text{m}\ (5)$$

In a working gain switched laser, application of the external field should increase the gain coefficient of the medium at least by an order of magnitude. It is clear from the estimates given above that we should look for transitions which meet the following requirements.

a) they are forbidden for M2 radiation (eq. (4));

b) they occur at wavelengths of 1 to 10 μm. (eqs. (3), (5)).

Increasing the wavelength by an order of magnitude will decrease the rate for E2 relative to E1 radiation by two orders and increase the rate of M1 relative to E2 radiation by the same factor. These requirements are met by vibrational transitions in homonuclear diatomic molecules.

Homonuclear molecules like H_2 do not possess a transition dipole moment for either vibrational or rotational transitions. Furthermore their magnetic dipole moment does not depend on the internuclear separation; hence vibrational transitions cannot occur for M1-radiation either. These transitions are observed however as electric quadrupole transitions, in Raman scattering and as field induced dipole transitions [5,6]. Vibrational transition with $\Delta v = \pm 1$ are strongest and are the only ones observed so far. Transition moments for $|\Delta v| > 1$ are found to be at least one order of magnitude smaller [7].

The rotational selection rules obeyed are:

$\Delta J = -2$ O-branch,
$\Delta J = 0$ Q-branch,
$\Delta J = +2$ S-branch.

For H_2 the transition between the first vibrational state and the groundstate occurs at a wavelength of $\lambda = 2.4$ μm. Hence we can expect the field induced transition rate to be at least one order of magnitude larger than the quadrupole rate.

As motivated above we now turn to a calculation of the field induced rate [2]. Let us first recall the physical mechanism of field induced transition in homonuclear molecules. The external field will induce a dipole moment in the electronic charge cloud. This induced moment will depend on the internuclear separation, and is hence coupled to the vibrational motion of the molecule. The rotational selection rules stem from the fact that in the transition one quantum of angular momentum is transferred to the photon emitted and one quantum is exchanged with the external field.

The square matrix element of the induced transition moment then is given by [6]:

$$\frac{1}{2J+1}\sum_{M,M'}|\langle vJM|D|v'J'M'\rangle|^2$$

$$= \left(\alpha_{01}^2 + \frac{4}{45}\frac{J(J+1)}{(2J+3)(2J-2)}\gamma_{01}^2\right)F^2 \qquad J' = J,$$

$$= \frac{2}{15}\frac{J(J+1)}{(2J+1)(2J-1)}\gamma_{01}^2F^2 \qquad J' = J-2$$

$$= \frac{2}{15}\frac{(J+2)(J+1)}{(2J+1)(2J+3)}\gamma_{01}^2F^2 \qquad J' = J+2.$$

Table 1
Experimental and theoretical results for the polarisability matrix elements of the molecules H_2 and N_2

	H_2		N_2	
	exp.	theor.	exp.	theor.
α_{01} (10^{-25} cm^3)	1.2 [6];	1.1 [15]	0.5 [16];	0.48, 0.38 [17]
γ_{01} (10^{-25} cm^3)	0.7 [14];	0.9 [15]	0.7 [16];	0.54, 0.38 [17]
λ^{-1} (10^3 cm^{-1})	4.16		2.36	

Here α_{01} and γ_{01} are matrix elements of the electronic groundstate polarisabilities $\alpha(R)$ and $\gamma(r)$ (R = internuclear distance) taken between the vibrational levels considered. α and γ are defined by

$$\alpha = \tfrac{2}{3}\alpha_\perp + \tfrac{1}{3}\alpha_\parallel, \qquad \gamma = \alpha_\parallel - \alpha_\perp,$$

where

$$\alpha_{xx} = \alpha_{yy} = \alpha_\perp, \qquad \alpha_{zz} = \alpha_\parallel$$

are the components of the electronic polarisability tensor. Transitions in the Q-branch ($\Delta J = 0$) are strongest. Neglecting contributions from γ_{01} which are of the same order of magnitude as probable errors in α_{01} we obtain for Q-transitions the spontaneous rates

$$W = \frac{32\pi^3}{h}\lambda^{-3}[\alpha_{01}F]^2,$$

$$= 0.9 \times 10^{-4}\ \mathrm{s}^{-1} \qquad \text{for } H_2,$$

$$= 3.4 \times 10^{-6}\ \mathrm{s}^{-1} \qquad \text{for } N_2,$$

both at $F = 10^5$ V/cm.

Although these rates are rather low we still anticipate reasonable gains since the field induced lines undergo collisional narrowing. This phenomenon occurs if the molecule undergoes many collisions while traveling over distances of about one wavelength [8]. As a result the lines are pressure broadened with width well below the doppler width at densities of 1 amagat.

Experimental and theoretical data on the collision narrowed linewidth are available for H_2. From ref. [6] we take the value of pressure broadening coefficient

$$\Delta(\lambda^{-1}) = 2.1 \times 10^{-3} \cdot N[\text{amagat}]\ \text{cm}^{-1}$$

The gain coefficient K is then given by

$$k = \Delta N \frac{\lambda^2}{4\pi^2}\frac{W}{c\Delta(\lambda^{-1})}$$

$$= \frac{\Delta N}{N}(5 \times 10^{-2})\ \text{cm}^{-1}$$

where $\Delta N/N$ is the relative population inversion. Keeping this quantity fixed the gain is independent of the density N in the pressure broadened regime, since the linewidth is proportional to N.

Higher gains can be obtained by increasing the strength of the applied field F. For a static field electric breakdown will occur at field strength larger than the value of 10^5 V/cm considered here. For the alternating electric field produced by a giant laser pulse much higher values are possible. For example a field strength of 10^5 V/cm would correspond to a laser intensity of 2×10^8 W/cm^2, but intensities of up to 10^{10} W/cm^2 have been achieved for picosecond pulses without optical breakdown [9].

In order to establish the feasibility of gain switching using a high power laser pulse as the source of our electric field, we use the following expression for the mixing of lasing and intermediate states a and r due to our switching laser signal [20]

$$|a(t)\rangle = |a\rangle + \left(\frac{\langle r|D\cdot F^{(+)}|a\rangle}{E_a - E_r + h\nu_0}\, e^{-i\omega_0 t}\right.$$

$$\left. + \frac{\langle r|D\cdot F^{(-)}|a\rangle}{E_a - E_r - h\nu_0}\, e^{i\omega_0 t}\right)|r\rangle, \qquad (6)$$

where $F^{(\pm)}$ denotes the positive and negative parts of the injected field, and ν_0 is the frequency of the injected field.

From eq. (6) we see that the injected laser field

mixes states in much the same way as does an ordinary dc field. However as noted above the fields are now much larger and this would increase the gain figures obtained above by at least two orders of magnitude. In this case gain figures of several percent cm^{-1} could be expected for the N_2 molecule also. For this molecule efficient vibrational pumping techniques such as electron impact [10] and discharges [11] are available.

From the semiclassical point of view, application of a time dependent electric field at frequency ν_0 will lead to an induced transition dipole moment oscillating at frequencies $\nu + \nu_0$ and $\nu - \nu_0$, where ν is the vibrational transition frequency. Hence stimulated emission will be observed at both sidebands. In a quantum mechanical treatment we would have to take into account the fact that the gain cross-section for both emission frequencies are not exactly equal [12,13,18].

In the present work the notion of inducing a dipole transition matrix element via a time-dependent laser field was an outgrowth of our calculations involving a dc electric field. Those calculations were based upon the earlier work of Overhauser [20]. Upon completing our analysis, it became apparent that there is a close connection with the earlier work of Harris et al., in which they utilized an incident laser to induce transitions from metastable atomic levels [18,19]. A more detailed discussion of the present work, connection with the work of Harris et al., and the question of population inversion in our H_2 system will be given elsewhere.

The authors gratefully acknowledge helpful discussions with P. Avizonis, K. Kompa and S. Rabin.

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Theory of time-delayed measurement: Subnatural linewidth and transient dip spectroscopy

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Time-delayed measurement of naturally broadened transitions can lead to a narrowing of the linewidth. Moreover, under appropriate conditions, it may result in the appearance of a dip at the line center. An analysis of time-delayed measurement thus provides a theoretical basis for useful optical techniques yielding high spectral resolution. Such an analysis is presented in this work.

I. INTRODUCTION

It has traditionally been one of the main endeavors of spectroscopists to develop measurement techniques yielding ever-higher resolution. In optical experiments the precision of the measurement is often limited by the broadening of the linewidth caused by the interaction of the system under investigation with its environment, such as Doppler effect, collisions, and spontaneous decay. It might seem that, after the Doppler- or collision-broadened width has been eliminated using one of the many schemes introduced in the past for that purpose,[1] the natural linewidth remains the ultimate limit to high-resolution spectroscopy.

Recently, we[2] have proposed and analyzed some spectroscopic techniques which provide resolution beyond the natural linewidth. These considerations are based on the fact[3] that, in the transient regime, the probability for induced transitions in a two-level system interacting with a monochromatic electromagnetic field is not weighted by a Lorentzian of width $\gamma_{ab} \equiv \gamma_a + \gamma_b$, but rather $\delta_{ab} \equiv \gamma_a - \gamma_b$. [$\gamma_a$ and γ_b are the decay rates of the two levels. Note that in this paper, we call γ_a and γ_b the amplitude (rather than population) decay rates. Thus γ_a and γ_b are twice as large as in the usual notation.] As a specific example to demonstrate such transient line narrowing, we have proposed an experimental setup, inspired from delayed detection level-crossing spectroscopy, that utilizes a time delay between the system preparation and the observation of emitted radiation.

Knight and Coleman[4] have shown that a similar result may be achieved in a system of two-level atoms weakly driven by an exponentially decaying laser pulse. In this system, although the lower level is stable, the exponential decay of the pulse amplitude has the same effect on the fluorescence spectrum as would the exponential decay of the lower level. Metcalf and Phillips[5] have shown that despite the loss of signal associated with time-delayed detection, it may still prove very useful in a number of applications. For example, as emphasized in Ref. 2, this technique would allow us to measure the difference $(\gamma_a - \gamma_b)$ directly, and therefore to a much higher precision than could be obtained from independent measurements of γ_a and γ_b.

We note that transient line-narrowing spectroscopy has a number of similarities with detection schemes developed earlier to achieve resolution beyond the natural linewidth in Mössbauer,[6,7] level-crossing,[8-11] and Lamb-shift[12-14] experiments. The common feature of these experiments is to discard the part of the radiation emitted shortly after the preparation of the system and to collect only the delayed and exponentially weakened signal.

In this paper we present a fully quantum-mechanical treatment of time-delayed spectroscopy, both for weak and strong incident fields. In the

discussion of Ref. 2 the atoms were assumed to be driven by a weak classical field, so that a perturbative treatment can be used. In general, the strong-field dynamics is substantially different from the weak-field one. Since the time-delayed spectrum depends sensitively on the temporal behavior of the system, one might expect that the inclusion of power broadening would lead to a different time-delayed spectrum. In fact, as is shown in the second part of this paper, it can lead to the appearance of a transient dip at line center. This dip at line center may prove to be a useful technique to determine accurately the position of the transition. Thus, time-delayed fluorescence measurements have the capability of providing high spectral resolution, not only through the line-narrowing effect, but possibly via strong-field "transient dip spectroscopy".

The goals of the present paper is threefold. First, we show that the results previously obtained semiclassically are recovered exactly in a fully quantum-mechanical treatment. Second, we extend the previous "weak-field" calculations to arbitrary strength driving fields. Third, we give an intuitive physical picture of transient line narrowing and transient dip spectroscopy, based on a well-known feature of the Rabi problem.

The remainder of this paper is organized in the following way: In Sec. II, we give a fully quantum-mechanical theory of transient line narrowing, considering an atom weakly driven by either cw or pulsed excitation. In the case of pulsed excitation, square pulses, as well as exponentially decreasing pulses, are considered. We show that this leads to exactly the same results as the semiclassical theory,[2-4] provided spontaneous emission directly between the two states under consideration is neglected, and the emitted photons from the two states are distinguishable. In Sec. III the effect of power broadening on the time-delayed spectrum is studied. It is shown that the inclusion of power broadening leads to the appearance of a transient dip. Finally, Sec. IV is a summary and discussion. Throughout the paper, natural units $\hbar = c = 1$ are used, unless otherwise stated.

II. QUANTUM-MECHANICAL THEORY OF TRANSIENT LINE NARROWING: WEAK-FIELD LIMIT

A. Four-level atom driven by weak cw radiation

We first consider the system previously investigated by Meystre, Scully, and Walther.[2] This system consists of an atom with two unstable levels a and b and a weak cw field driving the atom from the lower level b to the upper level a. If one includes the lower levels to which a and b decay, this may be considered as a four-level atom (see Fig. 1). We prepare the atom in level b and, as the field drives the atom to level a, we count the photons emitted following the $a \rightarrow c$ transition, starting a finite time θ after the atom is prepared. The counting rate is measured as a function of the detuning between the laser frequency k_0 and the energy separation Δ_{ab} between a and b.

Under the rotating-wave approximation and in the interaction picture, the wave function may be written as

$$|\psi(t)\rangle = \alpha^a(t)\ |a,(N-1)\vec{k}_0\rangle + \alpha^b(t)\ |b,N\vec{k}_0\rangle + \int d\vec{k}_1 \alpha^c_{\vec{k}_1}(t)\ |c,(N-1)\vec{k}_0,\vec{k}_1\rangle + \int d\vec{k}_2 \alpha^d_{\vec{k}_2}(t)\ |d,N\vec{k}_0,\vec{k}_2\rangle , \tag{2.1}$$

with

$$|\psi(0)\rangle = |b,N\vec{k}_0\rangle . \tag{2.2}$$

Here N is the number of photons in the driving field, $|a,(N-1)\vec{k}_0\rangle$ denotes the state in which the atom is in the state a and $(N-1)$ photons of mode $\vec{k}_0$ are present in the radiation field (and similarly for other eigenstates), and $\vec{k}_0$ is the wave vector of the driving field, $\vec{k}_1$ and $\vec{k}_2$ denote the modes of the photons emitted following the $a \rightarrow c$ and $b \rightarrow d$ transitions, respectively. In writing Eq. (2.1) we have assumed that the energy separations between any two levels are sufficiently different from one another that the photons $\vec{k}_0$, $\vec{k}_1$, and $\vec{k}_2$ are distinguishable. We also have assumed that spontaneous emission from a to b can be neglected. Substituting Eq. (2.1) into the Schrödinger equation, we im-

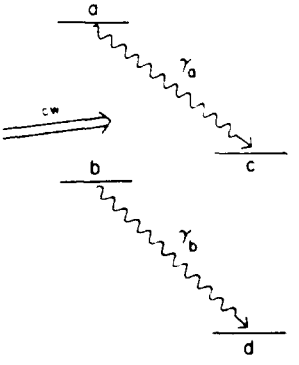

FIG. 1. Four-level atom driven by cw radiation.

mediately obtain the following equations for the probability amplitudes

$$i\frac{d}{dt}\alpha^a(t) = \sqrt{N}\,M^*_{k_0}\exp[i(\Delta_{ab}-k_0)t]\alpha^b(t) + \int d\vec{k}_1 M^*_{k_1}\exp[i(\Delta_{ac}-k_1)t]\alpha^c_{k_1}(t)\,, \tag{2.3a}$$

$$i\frac{d}{dt}\alpha^b(t) = \sqrt{N}\,M_{k_0}\exp[-i(\Delta_{ab}-k_0)t]\alpha^a(t) + \int d\vec{k}_2 M^*_{k_2}\exp[i(\Delta_{bd}-k_2)t]\alpha^d_{k_2}(t)\,, \tag{2.3b}$$

$$i\frac{d}{dt}\alpha^c_{k_1}(t) = M_{k_1}\exp[-i(\Delta_{ac}-k_1)t]\alpha^a(t)\,, \tag{2.3c}$$

$$i\frac{d}{dt}\alpha^d_{k_2}(t) = M_{k_2}\exp[-i(\Delta_{bd}-k_2)t]\alpha^b(t)\,, \tag{2.3d}$$

subject to the initial condition, Eq. (2.2), or

$$\alpha^b(0) = 1\,, \tag{2.4a}$$

$$\alpha^a(0) = \alpha^c_{k_1}(0) = \alpha^d_{k_2}(0) = 0\,. \tag{2.4b}$$

Δ_{ij} is the energy separation between the states i and j, k_i is the energy of a photon, i.e., $k_i = |\vec{k}_i|$, and M_{k_i} is the atomic-transition amplitude defined as

$$\langle j,\vec{k}_i|H_I|i,0\rangle = M_{k_i}\exp[-i(\Delta_{ij}-k_i)t]\,, \tag{2.5}$$

where $i = a$ or b and $j = c$ or d for the present case. H_I denotes the interaction Hamiltonian in the interaction picture.

In order to have an unambiguous definition of the time-delayed power spectrum, one needs to introduce a model detector into the problem.[7] In this paper, we consider the same scheme as in Ref. 2, i.e., we detect the total number of photons spontaneously emitted following the transition $a \to c$ as a function of the detuning Δ between the driving field and the atomic energy separation Δ_{ab}: $\Delta = k_0 - \Delta_{ab}$. We then define the time-delayed spectrum $N(\Delta,\theta)$ as

$$N(\Delta,\theta) = 2\gamma_a\int_\theta^\infty dt_1\,|\alpha^a(t_1)|^2\,, \tag{2.6a}$$

i.e., as the number of photocounts from time θ on. This can be reexpressed as the number of emitted photons at $t=\infty$ minus that at $t=\theta$,

$$N(\Delta,\theta) = \int d\vec{k}_1\,|\alpha^c_{k_1}(t=\infty)|^2 - \int d\vec{k}_1\,|\alpha^c_{k_1}(t=\theta)|^2\,. \tag{2.6b}$$

Thus, the time-delayed photocounts can be obtained by solving Eqs. (2.3) and substituting the solutions into Eqs. (2.6a) or (2.6b). Details of the calculation are shown in Appendix A for the weak-field limit. Here we only show the result

$$N(\Delta,\theta) = \frac{2N|M_{k_0}|^2\gamma_a}{\Delta^2+\delta_{ab}^2}\left[\frac{\exp(-2\gamma_a\theta)}{2\gamma_a} + \frac{\exp(-2\gamma_b\theta)}{2\gamma_b} - \frac{2\exp(-\gamma_{ab}\theta)}{\Delta^2+\gamma_{ab}^2}(\Delta\sin\Delta\theta - \gamma_{ab}\cos\Delta\theta)\right], \tag{2.7}$$

where $\delta_{ab} = \gamma_a - \gamma_b$, $\gamma_{ab} = \gamma_a + \gamma_b$, and γ_a and γ_b are the decay rates of levels a and b, respectively.

We note that in the limit $\theta \to 0$, Eq. (2.7) yields the usual Lorentzian of width γ_{ab}. However, if θ is sufficiently large, only the first or the second term in the large parentheses of Eq. (2.7) remains, depending upon which of γ_a and γ_b is largest. Thus, the dependence of $N(\Delta,\theta)$ on Δ is determined mainly by the Lorentzian prefactor, and the linewidth approaches the difference δ_{ab}. This is the transient line-narrowing effect. In fact, Eq. (2.7) is identical with the expression obtained earlier[2] using the density-matrix equations and a classical description of the field. Here, Eq. (2.7) is obtained using a fully quantum-mechanical approach. The normalized power spectrum $N(\Delta,\theta)$ is shown in Fig. 2 for $\gamma_a = 3$ and $\gamma_b = 1$, and in Fig. 3 for $\gamma_a = 1.01$ and $\gamma_b = 1$.

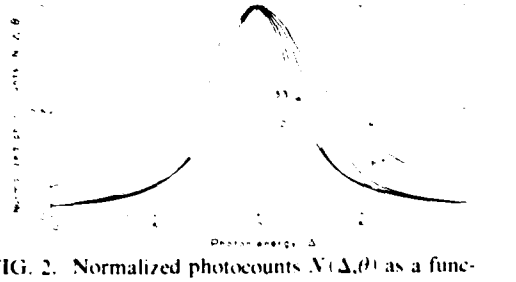

FIG. 2. Normalized photocounts $N(\Delta,\theta)$ as a function of Δ for various values of the delay $\theta = 0$, 0.67, 1.33, and 2.0 for the cases $\gamma_a = 3$ and $\gamma_b = 1$. θ is in units of γ_b^{-1}. If the system under consideration is the one shown in Fig. 1, Δ is the energy detuning between the driving field and the atomic-energy separation. If the system is that shown in Fig. 4, Δ is the energy detuning between the central frequency of the driving pulse and the atomic-energy separation, and γ_b is the decay constant of the exponentially decreasing pulse.

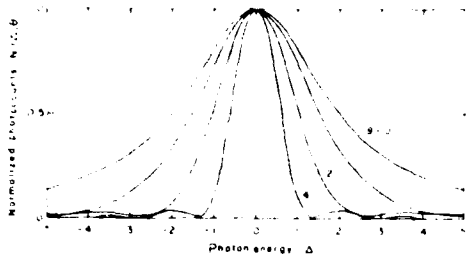

FIG. 3. Same as Fig. 2 except that $\theta = 0,1,2,4$ and $\gamma_a = 1.01$.

B. Three-level atom driven by weak-pulsed radiation

Knight and Coleman[4] have pointed out that transient line narrowing can also be realized in a system of two-level atoms driven by a weak exponentially decreasing pulse. In fact, the subnatural linewidth observed in Mössbauer experiments[6,7] involves basically the same system. For convenience we introduce, in addition to the two levels a and b, a third level c to which the upper level a can decay (see Fig. 4). Comparison of Figs. 1 and 4 shows the strong analogy between this system and that discussed previously. The only difference is that the exponentially decreasing pumping rate arises now from the exponential decay of the pulse amplitude in Fig. 4 whereas it came from the exponential decay of level b in Fig. 1.

We describe the radiation via its spectral amplitude $\phi(\vec{k})$. For an exponentially decaying pulse, this would be a Lorentzian. However, we keep $\phi(k)$ arbitrary, so that our result is valid for any pulse shape. Let us introduce the operator $B^\dagger$ as[16,17]

$$B^\dagger = \int d\vec{k}\phi(\vec{k})a_{\vec{k}}^\dagger , \tag{2.8}$$

where $a_{\vec{k}}^\dagger$ is the usual photon creation operator.

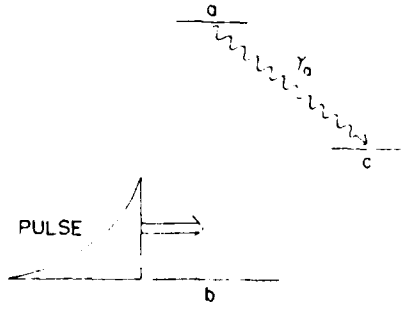

FIG. 4. Three-level atom driven by radiation pulse.

The pulsed radiation can then be represented by

$$|N_\phi\rangle = \frac{1}{\sqrt{N!}}(B^\dagger)^N|\text{vacuum}\rangle , \tag{2.9}$$

where N is the number of photons in the pulse. For simplicity, we take $N = 1$ and consistently limit ourselves to the weak-field limit.

As before, we write the wave function as

$$|\psi(t)\rangle = \alpha^a(t)|a,0\rangle + \int d\vec{k}\alpha_{\vec{k}}^b(t)|b,\vec{k}\rangle + \int d\vec{k}_1\alpha_{\vec{k}_1}^c(t)|c,\vec{k}_1\rangle , \tag{2.10}$$

with

$$|\psi(0)\rangle = \int d\vec{k}\phi(\vec{k})|b,\vec{k}\rangle = |b,1_\phi\rangle . \tag{2.11}$$

Substituting Eq. (2.10) into the Schrödinger equation, we obtain

$$i\frac{d}{dt}\alpha^a(t) = \int d\vec{k}M_{\vec{k}}^*\exp[i(\Delta_{ab}-k)t]\alpha_{\vec{k}}^b(t) + \int d\vec{k}_1 M_{\vec{k}_1}^*\exp[i(\Delta_{ac}-k_1)t]\alpha_{\vec{k}_1}^c(t) , \tag{2.12a}$$

$$i\frac{d}{dt}\alpha_{\vec{k}}^b(t) = M_{\vec{k}}\exp[-i(\Delta_{ab}-k)t]\alpha^a(t) , \tag{2.12b}$$

$$i\frac{d}{dt}\alpha_{\vec{k}_1}^c(t) = M_{\vec{k}_1}\exp[-i(\Delta_{ac}-k_1)t]\alpha^a(t) . \tag{2.12c}$$

where, as before, we have assumed that Δ_{ab} and Δ_{ac} are sufficiently different from each other so that the photons $\vec{k}$ and $\vec{k}_1$ are distinguishable. Equations (2.12) are subject to the initial condition, Eq. (2.11), or

$$\alpha_{\vec{k}}^b(0) = \phi(\vec{k}),\quad \alpha^a(0) = \alpha_{\vec{k}_1}^c(0) = 0 . \tag{2.13}$$

The number of photons emitted following the $a \to c$ transition from the time $t = \theta$ on is given by

$$N(\Delta,\theta) = 2\gamma_a\int_\theta^\infty dt_1\,|\alpha^a(t_1)|^2 = \int d\vec{k}_1\,|\alpha_{\vec{k}_1}^c(t=\infty)|^2 - \int d\vec{k}_1\,|\alpha_{\vec{k}_1}^c(t=\theta)|^2 . \tag{2.14}$$

This can be obtained by solving Eqs. (2.12) and substituting the solution into Eq. (2.14). Details of the calculation are shown in Appendix B, neglecting spontaneous decay from a to b (γ_a is the decay rate of the $a \to c$ transition). $N(\Delta,\theta)$, of course, depends on the pulse shape because the solutions of Eqs. (2.12) are different for a different choice of $\phi(\vec{k})$.

For the case of an exponentially decaying pulse, $N(\Delta,\theta)$ is again given by Eq. (2.7). Therefore,

Figs. 2 and 3 also give the normalized count $N(\Delta,\theta)$ for the system considered here. We note, however, that k_0 is now to be interpreted as the central frequency of the pulse, $\Delta = k_0 - \Delta_{ab}$, and γ_b is the decay constant of the pulse rather than of the atomic level.

For the case of a square pulse of duration t_0, we obtain

$$N(\Delta,\theta) = \begin{cases} \dfrac{2|g_s|^2\gamma_a}{\Delta^2+\gamma_a^2}\left[t_0-\theta-\dfrac{\exp(-2\gamma_a t_0)-\exp(-2\gamma_a\theta)}{2\gamma_a} + \dfrac{2\exp(-\gamma_a t_0)[\gamma_a\cos\Delta t_0-\Delta\sin\Delta t_0]-2\exp(-\gamma_a\theta)[\gamma_a\cos\Delta\theta-\Delta\sin\Delta\theta]}{\Delta^2+\gamma_a^2}\right] & \text{for } \theta < t_0, \quad (2.15a) \\ \dfrac{|g_s|^2\exp(-2\gamma_a\theta)}{\Delta^2+\gamma_a^2}[1+\exp(2\gamma_a t_0)-2\exp(\gamma_a t_0)\cos\Delta t_0] & \text{for } \theta \geq t_0, \quad (2.15b) \end{cases}$$

g_s is a constant depending on the pulse parameters, k_0 is again the central frequency of the pulse, and $\Delta = k_0 - \Delta_{ab}$. The normalized count $N(\Delta,\theta)$ is shown in Fig. 5 for $t_0 = 1$ and $\gamma_a = 3$, and in Fig. 6 for $t_0 = 1$ and $\gamma_a = 1.01$. $N(\Delta,\theta)$ takes the same form for all θ's exceeding t_0. This is because, for $\theta > t_0$, the situation is analogous to making time-delayed measurement on a two-level system with the levels a and c. We see that the line narrowing does occur with a square pulse although the narrowing is not as dramatic as that associated with an exponentially decreasing pulse.

C. Physical interpretation of transient line narrowing

Since transient line narrowing arises from the behavior of the system in the transient regime, it is natural to study the temporal behavior of the system in order to better understand and interpret the effect. Intuitively, it is not difficult to understand the line-narrowing effect. It is based on the well-known (but perhaps anti-intuitive) fact that the Rabi frequency is larger for larger detunings, and that therefore the excitation and the depletion of the population of the upper level (level a in our system) are slower when excited exactly on resonance. This means that the remaining population after some delay time θ is relatively a large number when excited on resonance, thus leading to the line narrowing. In this section we show quantitatively that the above interpretation is indeed true. Although our discussion here is limited to the case of an atom driven by pulsed radiation, it can equally be applied to the system considered in Sec. II A provided that the appropriate redefinition of k_0, γ_b, etc., is made.

The integrand $|\alpha^a(t)|^2$ in Eq. (2.14) for the photocount is the probability for the atom to be in the upper state a at time t. For the case of an exponentially decreasing pulse, we find using Eq. (B4) of Appendix B,

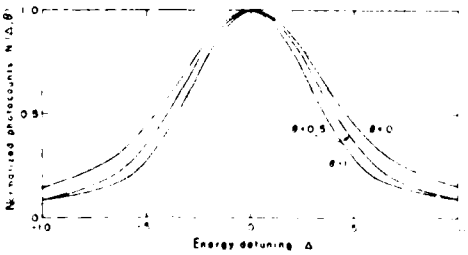

FIG. 5. Normalized photocounts $N(\Delta,\theta)$ as a function of energy detuning for various values of the delay $\theta = 0$, 0.5, and $\theta > 1$ for the case $\gamma_a = 3$. The driving field is assumed to be in the form of a square pulse of duration $t_0 = 1$. θ is in units of t_0.

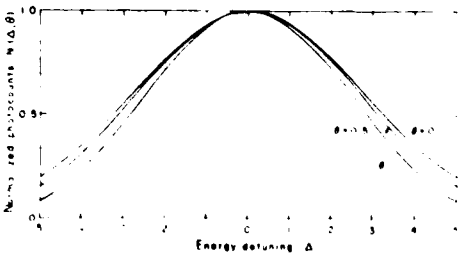

FIG. 6. Same as Fig. 5 except that $\gamma_a = 1.01$

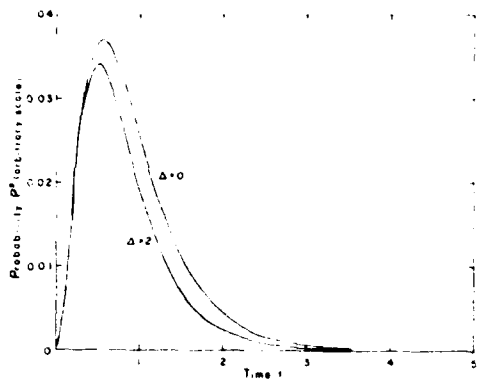

FIG. 7. Probability $P^a(\Delta,t)$ as a function of time t for two values of energy detuning $\Delta=0$ and 2 for the cases $\gamma_a=3$ and $\gamma_b=1$ (exponentially decreasing pulse). The time is in units of γ_b^{-1}.

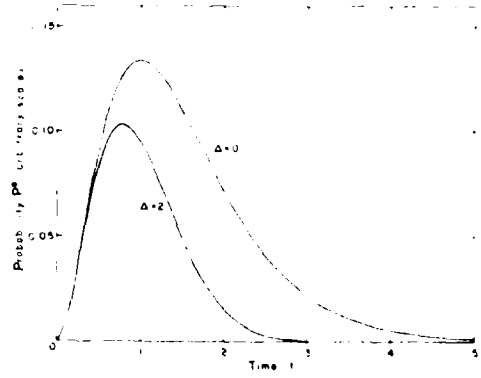

FIG. 8. Same as Fig. 7 except that $\gamma_a=1.01$.

$$p^a(\Delta,t)\equiv|\alpha^a(t)|^2 = \frac{|g_e|^2}{\Delta^2+\delta_{ab}^2}[\exp(-2\gamma_a t)+\exp(-2\gamma_b t) - 2\exp(-\gamma_{ab}t)\cos\Delta t]\,. \quad (2.16)$$

$p^a(\Delta,t)$ is plotted in Fig. 7 as a function of time for two different values of detuning $\Delta=0$ and 2 for the case $\gamma_a=3$ and $\gamma_b=1$, and in Fig. 8 for $\gamma_a=1.01$ and $\gamma_b=1$. $p^a(\Delta,t)$ initially increases as the driving field pumps the atom to level a, goes through a peak and eventually decreases to zero because of spontaneous emission. The peak occurs earlier and has a smaller value for $\Delta=2$. This reflects the above-mentioned fact that the Rabi frequency is larger for larger detunings and is the key point in understanding the line-narrowing effect. According to Eq. (2.14), $N(\Delta,\theta)$ is the area under the probability curve between $t=\theta$ and ∞. We see from Fig. 7 that, as θ is increased, this area decreases faster for larger Δ. This is even more clear in Fig. 8. The integral of $p^a(\Delta,t)$ between, for example, $t=2$ and ∞ is only a small fraction of the area between $t=0$ and ∞ for the case $\Delta=2$, while it is still a large fraction for the case $\Delta=0$. Thus, the larger Δ, the faster the system emits spontaneously and leaves a smaller number of photons to be emitted after some delay time θ. This directly leads to the narrowing of the linewidth.

For a square pulse of duration t_0, the probability takes the form

$$p^a(\Delta,t)=\begin{cases} |\alpha^a(t)|^2=\dfrac{|g_s|^2}{\Delta^2+\gamma_a^2}[1+\exp(-2\gamma_a t)-2\exp(-\gamma_a t)\cos\Delta t], & \text{for } t\le t_0 \quad (2.17a)\\ |\alpha^a(t)|^2=\dfrac{|g_s|^2\exp(-2\gamma_a t)}{\Delta^2+\gamma_a^2}[\exp(2\gamma_a t_0)+1-2\exp(\gamma_a t_0)\cos\Delta t_0], & \text{for } t>t_0 \quad (2.17b)\end{cases}$$

where Eqs. (B8) of Appendix B have been used. This probability is plotted in Fig. 9 as a function of time for two different values of detuning for the case $t_0=1$, $\gamma_a=1.01$. Here again we see that the peak occurs earlier and has a smaller value for larger detunings, although the effect is not as strong as before. This explains a relatively weak line narrowing with a square pulse.

III. STRONG-SIGNAL REGIME: TRANSIENT DIP SPECTROSCOPY

Up to now, we have restricted ourselves to the weak-field limit, in which the use of a perturbative treatment is justified. We now relax this restriction and study time-delayed spectroscopy in the strong-signal regime.

The analysis departs from that of the weak-field limit in that instead of using Eq. (A5) of Appendix A, we now solve Eqs. (A2) and (A3) exactly for $\alpha^a(t)$ and $\alpha^b(t)$. These equations yield a second-order differential equation for $\alpha^a(t)$ [and also for $\alpha^b(t)$], with the solution[3]

$$\alpha^a(t) = -i\sqrt{N} M^*_{\vec{k}_0} \exp(u_2 t) \int_0^t dt_1 \exp[(u_1 - u_2)t_1] , \tag{3.1}$$

where

$$u_1 = \{ -\gamma_{ab} - i\Delta + [(\delta_{ab} + i\Delta)^2 - 4N | M_{\vec{k}_0} |^2]^{1/2} \}/2 , \tag{3.2a}$$

$$u_2 = \{ -\gamma_{ab} - i\Delta - [(\delta_{ab} + i\Delta)^2 - 4N | M_{\vec{k}_0} |^2]^{1/2} \}/2 . \tag{3.2b}$$

Using Eqs. (3.1) and (2.3c) we get

$$\alpha^c_{\vec{k}_1}(t) = (-i)^2 \sqrt{N} M^*_{\vec{k}_0} M_{\vec{k}_1} \int_0^t dt_1 \exp[u_2 - i(\Delta_{ac} - k_1)t_1] \int_0^{t_1} dt_2 \exp(u_1 - u_2)t_2 . \tag{3.3}$$

The number of photons emitted during the time interval (θ, ∞) following the $a \rightarrow c$ transition is again given by Eq. (2.6a) or (2.6b). After tedious but straightforward algebra, we obtain

$$N(\Delta,\theta) = [4\gamma_a N | M_{\vec{k}_0} |^2 \exp(-\gamma_{ab}\theta)]/\rho \times \left| \frac{\gamma_{ab} \cosh[\theta\sqrt{\rho}\cos(\phi/2)] + \sqrt{\rho}\cos(\phi/2)\sinh[\theta\sqrt{\rho}\cos(\phi/2)]}{\gamma_{ab}^2 - \rho\cos^2(\phi/2)} - \frac{\gamma_{ab}\cos[\theta\sqrt{\rho}\sin(\phi/2)] - \sqrt{\rho}\sin(\phi/2)\sin[\theta\sqrt{\rho}\sin(\phi/2)]}{\gamma_{ab}^2 + \rho\sin^2(\phi/2)} \right| , \tag{3.4}$$

where

$$\rho = [(\delta_{ab}^2 - \Delta^2 - 4N | M_{\vec{k}_0} |^2)^2 + 4\delta_{ab}^2\Delta^2]^{1/2} , \tag{3.5}$$

and ϕ is determined by

$$\cos\phi = (\delta_{ab}^2 - \Delta^2 - 4N | M_{\vec{k}_0} |^2)/\rho , \tag{3.6a}$$

$$\sin\phi = 2\delta_{ab}\Delta/\rho . \tag{3.6b}$$

Equation (3.4) is the main result of this section. The time-delayed count $N(\Delta,\theta)$ is, in general, a complicated function of the system parameters, and we first consider some limiting cases.

A. Weak-field limit

In the weak-field limit $N \rightarrow 0$, we obtain

$$\rho \simeq \delta_{ab}^2 + \Delta^2 , \tag{3.7a}$$

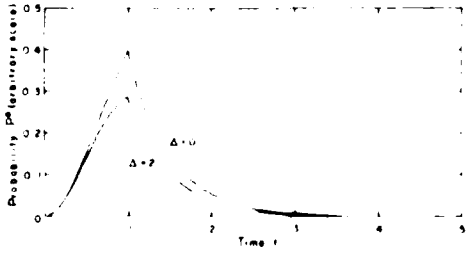

FIG. 9. Probability P^a as a function of time t for two values of energy detuning $\Delta = 0$ and 2 for the case $t_0 = 1$ (square pulse) and $\gamma_a = 1.01$. t is in units of t_0.

$$\cos\phi \simeq (\delta_{ab}^2 - \Delta^2)/\rho , \tag{3.7b}$$

$$\sin\phi \simeq 2\delta_{ab}\Delta/\rho . \tag{3.7c}$$

Equation (3.4) reduces then to Eq. (2.7) of Sec. II A, as it should.

B. No time delay

In the limit $\theta \rightarrow 0$ (no time delay), Eq. (3.4) becomes

$$N(\Delta,\theta=0) = \frac{N | M_{\vec{k}_0} |^2 \gamma_{ab}}{\gamma_b(\Delta^2 + \gamma_{ab}^2 + N | M_{\vec{k}_0} |^2 \gamma_{ab}^2/\gamma_a\gamma_b)} , \tag{3.8}$$

which is the well-known power-broadened Lorentzian line shape.

C. General case

We now return to the general formula Eq. (3.4). In Figs. 10 and 11, $N(\Delta,\theta)$ is plotted as a function of the detuning $\Delta(\equiv k_0-\Delta_{ab})$ for different delay times θ and for different field intensities for the case $\gamma_a=3$ and $\gamma_b=1$. We immediately see a new feature of the power-broadened spectrum, namely, the appearance of a dip at line center for large θ and for high enough field intensities. A close investigation indicates that, for a fixed value of the field intensity, and as θ is increased from zero, the linewidth first decreases (transient line narrowing). However, as θ is further increased, the line broadens back until the linewidth becomes roughly the same as that at $\theta=0$. If θ is further increased beyond this point, a narrow dip appears at line center, and becomes deeper and wider as θ is further increased. Since it always appears at line center, it may help locate the center of a transition with improved accuracy over standard methods. The dip appears with a smaller delay for stronger fields. Thus, one can operate without significant loss of signal. For example, for a Rabi frequency $G(\equiv N\,|M_{\vec{k}_0}|^2)=1.5$, the dip appears already at $\theta=2$. On the other hand, for $G=0.6$, the dip still does not appear even for θ as large as 4. In Fig. 12 we show $N(\Delta,\theta)$ at a fixed θ as G is varied. Here again, we see clearly that the dip appears more easily for stronger fields.

In Fig. 13 we show the same $N(\Delta,\theta)$ versus Δ curve for the cases $\gamma_a=1.01$ and $\gamma_b=1$, and $G=1$ for various θ. We immediately notice the strong oscillatory behavior exhibited in this case.

A useful parameter to determine the behavior of $N(\Delta,\theta)$ is the ratio of the Rabi frequency to δ_{ab}, or

$$r\equiv G^2/\delta_{ab}^2 \ . \tag{3.9}$$

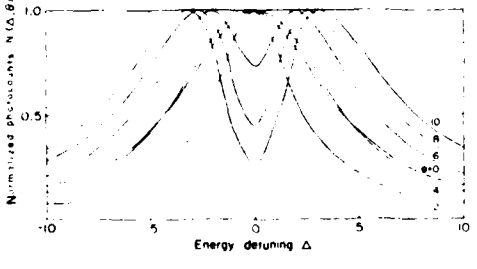

FIG. 10. Normalized photocounts $N(\Delta,\theta)$ as a function of energy detuning Δ for various values of the delay $\theta=0,2,4,6,8,10$ for the case $\gamma_a=3$, $\gamma_b=1$, and the Rabi frequency $G=N\,|M_{\vec{k}_0}|^2=0.6$. θ is in units of γ_b^{-1}.

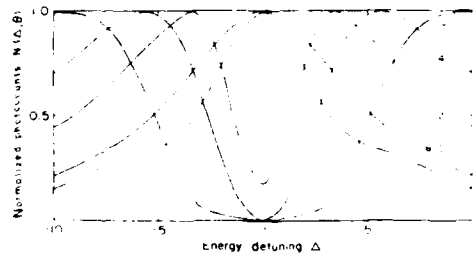

FIG. 11. Same as Fig. 10 except that $\theta=0,1,2,4,10$ and $G=1.5$.

This ratio determines whether the arguments of the hyperbolic functions are larger or smaller than those of the sinusoidal functions in Eq. (3.4), and therefore determines the characteristic behavior of $N(\Delta,\theta)$. For the case of Fig. 13, r is very large ($r=10^4$) and the contribution from the sinusoidal terms is important. As a result $N(\Delta,\theta)$ exhibits a strong oscillatory behavior. On the other hand, r is relatively small ($r<1$) for the cases shown in Figs. 10 and 11, and no strong oscillatory behavior is exhibited there.

In a sense the transient dip is an opposite phenomenon to the line narrowing because, in order for the dip to appear, the remaining population of the upper level a after the delay time θ should be a relatively small number when excited on resonance. That is, the depletion of the population of level a should be faster when excited on resonance. That this is indeed the case when θ is sufficiently large can be seen in Fig. 14, where the probability $p^a(\Delta,t)\equiv|\alpha^a(t)|^2$ for the atoms to be in level a is plotted as a function of time for two different values of detuning for the cases $\gamma_a=3$, $\gamma_b=1$, and $G=0.6$. As we have already noted in the previous section, the peak occurs earlier and has a smaller

FIG. 12. Normalized photocounts $N(\Delta,\theta)$ as a function of energy detuning Δ for various values of the field intensity $G=0.3$, 0.6, 0.9, and 1.5 for the cases $\gamma_a=3$, $\gamma_b=1$, and for a fixed delay time $\theta=4$. θ is in units of γ_b^{-1}.

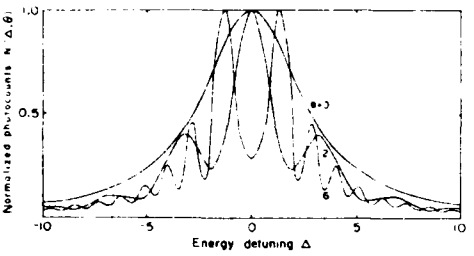

FIG. 13. Normalized photocounts $N(\Delta,\theta)$ as a function of energy detuning Δ for various values of the delay $\theta=0,2,6$ for the cases $\gamma_a=1.01$, $\gamma_b=1$, and $G=1$. θ is in units of γ_b^{-1}.

value for $\Delta=2$. This of course accounts for the narrowing of the linewidth for small values of the delay time θ. However, when θ is sufficiently large, the $\Delta=2$ curve finally catches up with the $\Delta=0$ curve. This behavior can be understood if we note that for a large detuning the atom is inefficiently pumped to level a and therefore still a large number of atoms are available to be pumped at a large time. This means a slower depletion of level a at sufficiently large time when excited off resonance and leads directly to the appearance of a dip at line center.

IV. SUMMARY AND DISCUSSION

Under appropriate conditions, time-delayed observation of the radiation emitted during an atomic transition can lead to the narrowing of the linewidth or the appearance of a dip at line center. The linewidth is limited in the small-signal regime by the difference in the transition rates involved. This provides an optical technique that yields high spectral resolution.

A dip may appear at line center as a result of the transient behavior of a system subject to power broadening. Unlike the line-narrowing effect, the width of the dip is not limited by $(\gamma_a-\gamma_b)$, and/or the delay θ. It can be as small as one likes if the intensity of the driving field is close to the threshold for its appearance. This dip suggests a means of enhancing resolution involving homogeneously broadened systems while the Lamb dip is available to study inhomogeneously broadened systems.

The narrowing of the linewidth or the appearance of the dip is achieved at the expense of some loss of the signal. A recent analysis[5] indicates that, despite the signal loss, time-resolved line narrowing is highly desirable in a large number of cases. If one tries to locate line center in the absence of spectral complications, i.e., if we know we have only one line in the region of interest, then it may be best to let the delay time $\theta\to 0$ and collect the maximum number of counts, since the determination of the line center improves as the square root of the intensity of the signal. However, if there are complicating circumstances, such as overlapping lines, etc., it may be better to use finite delay times with the attendant line narrowing and/or dip.

We finally note that the results obtained here via a fully quantum-mechanical theory take exactly the same form as those previously derived semiclassically. The origin of this exact agreement lies in the neglect of spontaneous decay from level $|a\rangle$ to $|b\rangle$.

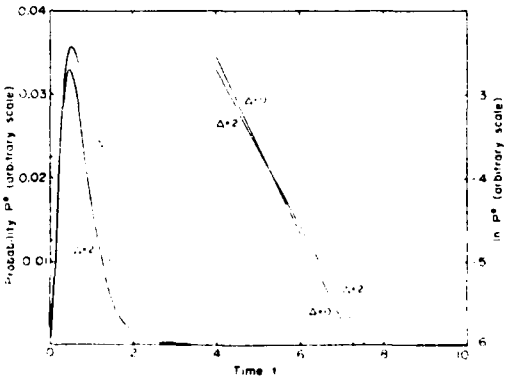

FIG. 14. Probability P^a as a function of time t for two values of energy detuning $\Delta=0$ and 2, for the case $\gamma_a=3$, $\gamma_b=1$, and $G=0.6$. t is in units of γ_b^{-1}. For $t\geq 4$, $\ln P^a$ is plotted as a function of time t and the corresponding scale is shown on the right side of the graph.

ACKNOWLEDGMENT

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APPENDIX A: DERIVATION OF EQ. (2.7). THE CASE OF cw RADIATION

In order to derive Eq. (2.7), we first solve Eqs. (2.3). From Eq. (2.3c) we have

$$\alpha^c_{k_1}(t)=-iM_{k_1}\int_0^t dt_1\exp[-i(\Delta_{ac}-k_1)t_1]\alpha^a(t_1)\,. \tag{A1}$$

Substituting Eq. (A1) into Eq. (2.3a), we obtain

$$\left[\frac{d}{dt}+\gamma_a\right]\alpha^a(t)=-i\sqrt{N}M^*_{\vec{k}_0}\exp(-i\Delta t)\alpha^b(t)\ . \tag{A2}$$

Similarly, from Eqs. (2.3d) and (2.3b), we obtain

$$\left[\frac{d}{dt}+\gamma_b\right]\alpha^b(t)=-i\sqrt{N}M_{\vec{k}_0}\exp(i\Delta t)\alpha^a(t)\ . \tag{A3}$$

The decay rates γ_a and γ_b in Eqs. (A2) and (A3) are defined as

$$\gamma_a=\left[k^2\pi\int d\Omega_{\hat{k}}\,|M_{\vec{k}}|^2\right]_{k=\Delta_{ac}}\ , \tag{A4}$$

γ_b is given by the same expression except that it is evaluated at $k=\Delta_{bd}$ instead of $k=\Delta_{ac}$.

In the weak-field limit (N small, or Rabi frequency $G<<\gamma_a,\gamma_b$), the right side of Eq. (A3) may be neglected. This means that the major source of depleting level b is not the driving field but spontaneous decay to level d. We then have

$$\alpha^b(t)\cong\exp(-\gamma_b t)\ . \tag{A5}$$

Substituting Eq. (A5) into (A2) we then obtain

$$\alpha^a(t)\cong -i\sqrt{N}M^*_{\vec{k}_0}\exp(-\gamma_a t)\int_0^t dt_1\exp[(-\delta_{ab}-i\Delta)t_1] \tag{A6}$$

and therefore, using Eq. (A1),

$$\alpha^c_{\vec{k}_1}(t)\cong(-i)^2\sqrt{N}M^*_{\vec{k}_0}M_{\vec{k}_1}\int_0^t dt_1\exp[-\gamma_a-i(\Delta_{ac}-k_1)]t_1\int_0^{t_1}dt_2\exp[(-\delta_{ab}-i\Delta)t_2]\ . \tag{A7}$$

Substituting Eq. (A6) into (2.6a), or Eq. (A7) into (2.6b), we immediately obtain Eq. (2.7) for the time-delayed photocounts.

APPENDIX B: DERIVATION OF EQS. (2.7) AND (2.15). THE CASE OF PULSED RADIATION

In order to derive Eqs. (2.7) and (2.15) for the case of pulsed radiation, we first solve Eqs. (2.12). From Eqs. (2.12b) and (2.12c), we have

$$\alpha^b_{\vec{k}}(t)=\phi(\vec{k})-iM_{\vec{k}}\int_0^t dt_1\exp[-i(\Delta_{ab}-k)t_1]\alpha^a(t_1)\ , \tag{B1}$$

$$\alpha^c_{\vec{k}_1}(t)=-iM_{\vec{k}_1}\int_0^t dt_1\exp[-i(\Delta_{ac}-k_1)t_1]\alpha^a(t_1)\ . \tag{B2}$$

Substituting Eqs. (B1) and (B2) into Eq. (2.12a), we obtain

$$\left[\frac{d}{dt}+\gamma_a+\gamma'_a\right]\alpha^a(t)$$

$$=-i\int d\vec{k}M^*_{\vec{k}}\exp[i(\Delta_{ab}-k)t]\phi(\vec{k})\ , \tag{B3a}$$

$$=-ig(t)\ , \tag{B3b}$$

where γ_a and γ'_a are the spontaneous decay rates from level a to c and b, respectively, and Eq. (B3b) defines the pulse-shape function $g(t)$.[16,17] From here on we assume that γ'_a is negligibly small.

From Eq. (B3b) we then have

$$\alpha^a(t)=-i\exp(-\gamma_a t)\int_0^t dt_1\exp(\gamma_a t_1)g(t_1)\ , \tag{B4}$$

and from Eq. (B2) we have

$$\alpha^c_{\vec{k}_1}(t)=(-i)^2M_{\vec{k}_1}\int_0^t dt_1\exp[-\gamma_a-i(\Delta_{ac}-k_1)]t_1\times\int_0^{t_1}dt_2\exp(\gamma_a t_2)g(t_2)\ . \tag{B5}$$

The time-delayed photocounts $N(\Delta,\theta)$ can be obtained by substituting Eq. (B4) or (B5) into Eq. (2.14). $N(\Delta,\theta)$ depends on the pulse shape through the function $g(t)$.

For an exponentially decreasing pulse, the pulse-shape function $g(t)$ takes the form

$$g(t)=g_e\exp[(-\gamma_b-i\Delta)t]\ , \tag{B6}$$

where g_e is a constant, $\Delta=k_0-\Delta_{ab}$, k_0 is the central frequency of the pulse, and γ_b is the decay constant of the pulse. Substituting Eq. (B6) into Eq. (B4) or (B5), and using Eq. (2.14), we obtain Eq. (2.7) for the time-delayed count $N(\Delta,\theta)$.

For a square pulse of duration t_0, we have

$$g(t)=\begin{cases}g_s\exp(-i\Delta t), & 0\le t\le t_0\\ 0, & t>t_0\end{cases} \tag{B7}$$

where g_s is a constant, k_0 is again the central frequency of the pulse, and $\Delta=k_0-\Delta_{ab}$. Substituting Eq. (B7) into Eqs. (B4) and (B5), we obtain

$$\alpha^a(t)=\begin{cases} -ig_s \exp(-\gamma_a t)\int_0^t dt_1 \exp[(\gamma_a - i\Delta)t_1], \quad 0\le t\le t_0 & \text{(B8a)} \\ -ig_s \exp(-\gamma_a t)\int_0^{t_0} dt_1 \exp[(\gamma_a - i\Delta)t_1], \quad t > t_0 & \text{(B8b)} \end{cases}$$

and, for $t > t_0$,

$$\alpha^c_{k_1}(t)=(-i)^2 g_s M_{k_1}\left\{\int_0^{t_0} dt_1 \exp[-\gamma_a - i(\Delta_{ac}-k_1)]t \int_0^{t_1} dt_2 \exp[(\gamma_a - i\Delta)t_2]\right.$$
$$\left. + \int_{t_0}^{t} dt_1 \exp[-\gamma_a - i(\Delta_{ac}-k_1)]t_1 \int_0^{t_0} dt_2 \exp[(\gamma_a - i\Delta)t_2]\right\}. \qquad \text{(B9)}$$

Substituting Eq. (B8) or (B9) into Eq. (2.14), we obtain Eqs. (2.15) for the time-delayed photocounts.

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RADIATIVE ENERGY TRANSFER IN A RANDOMLY FLUCTUATING MEDIUM*

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The basic laws of the phenomenological theory of radiative energy transfer are derived, under certain conditions, within the framework of the stochastic scalar wave theory. An equation of radiative energy transfer is derived for wave propagation in a statistically quasihomogeneous medium. Our results relate the extinction and scattering coefficients (which are introduced heuristically in the conventional theory of radiative energy transfer) to the stochastic characteristics of the medium.

1. Introduction

The subject of radiative energy transfer through stellar atmospheres and through turbulent media is generally treated on the basis of a phenomenological theory [1,2]. In recent years, many attempts have been made towards providing a satisfactory basis for the conventional theory and to deliminate its range of validity [3–14]. These attempts have met so far with a limited success.

The central quantity of the phenomenological theory is the so-called specific intensity of radiation $I(\boldsymbol{R},\boldsymbol{s})$ which satisfies the radiative energy transfer equation of the form

$$\boldsymbol{s}\cdot\nabla_R I(\boldsymbol{R},\boldsymbol{s}) = -\alpha(\boldsymbol{R},\boldsymbol{s})\, I(\boldsymbol{R},\boldsymbol{s}) + \int \beta(\boldsymbol{R},\boldsymbol{s},\boldsymbol{s}')\, I(\boldsymbol{R},\boldsymbol{s}')\, \mathrm{d}\Omega_{s'} + D(\boldsymbol{R},\boldsymbol{s}). \tag{1.1}$$

In this equation the left-hand side represents the rate of change of the specific intensity along the $\boldsymbol{s}$-direction. The first term on the right represents "extinction" due to absorption and scattering, the second term represents a contribution due to scattering from all directions (the integration extending over the complete 4π-solid angle generated by the unit vector $\boldsymbol{s}'$) and the last term represents the effect of sources. The functions $\alpha(\boldsymbol{R},\boldsymbol{s})$, $\beta(\boldsymbol{R},\boldsymbol{s},\boldsymbol{s}')$ and $D(\boldsymbol{R},\boldsymbol{s})$ are the so-called extinction coefficient, the differential scattering coefficient and the source function respectively.

In the usual heuristic model, the specific intensity $I(\boldsymbol{R},\boldsymbol{s})$ is treated as a radiometric quantity and the equation of radiative energy transfer is derived by an intuitive quasi-geometrical argument involving balance of radiant energy, without elucidating the microscopic meaning of the extinction and scattering coefficients and of the source function.

In this paper we investigate the foundations of the theory of radiative energy transfer and derive an equation of radiative energy transfer in a randomly fluctuating medium on the basis of scalar wave theory. Our results elucidate the relationship between the extinction and scattering coefficients and the stochastic characteristic of the medium.

2. Relationship between the specific intensity and the coherence function of the wavefield

We consider a random scalar wavefield $v(\boldsymbol{r})$. The second-order-coherence function of the wavefield is given by

$$\Gamma(\boldsymbol{r}_1,\boldsymbol{r}_2) = \langle v(\boldsymbol{r}_1)\, v^*(\boldsymbol{r}_2)\rangle. \tag{2.1}$$

In eq. (2.2), the sharp brackets denote the ensemble

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average. The coherence function $\bar{\Gamma}(\boldsymbol{r}_1,\boldsymbol{r}_2)$, when expressed in terms of the new variables

$$\boldsymbol{R} = \tfrac{1}{2}(\boldsymbol{r}_1 + \boldsymbol{r}_2); \quad \boldsymbol{r} = \boldsymbol{r}_1 - \boldsymbol{r}_2, \tag{2.2}$$

will be denoted by $\Gamma(\boldsymbol{R},\boldsymbol{r})$; i.e.,

$$\Gamma(\boldsymbol{R},\boldsymbol{r}) \equiv \bar{\Gamma}(\boldsymbol{r}_1,\boldsymbol{r}_2). \tag{2.3}$$

The ensemble average of the energy density $U(\boldsymbol{R})$ and the ensemble average of the energy flux $\boldsymbol{F}(\boldsymbol{R})$ are related to the coherence function of the wavefield by the following equations [7]:

$$U(\boldsymbol{R}) = (1/c)\Gamma(\boldsymbol{R},0), \quad \boldsymbol{F}(\boldsymbol{R}) = (1/ik_0)[\nabla_{\boldsymbol{r}}\Gamma(\boldsymbol{R},\boldsymbol{r})]_{\boldsymbol{r}=0}, \tag{2.4),(2.5}$$

where c denotes the speed of light in vacuo and k_0 is the wavenumber in free space.

Not long ago, Wolf [12] formulated a rigorous theory of radiative transfer in free electromagnetic field. In his theory, two quantities are introduced which relate to energy transport; the so-called "angular components" of the average electromagnetic energy density and of the average Poynting vector. In analogy with these quantities, we introduce the angular component of the average energy density $Q(\boldsymbol{R},\boldsymbol{s})$ and the angular component of the average energy flux $\boldsymbol{T}(\boldsymbol{R},\boldsymbol{s})$ by the following relations:

$$U(\boldsymbol{R}) = \int Q(\boldsymbol{R},\boldsymbol{s})\,\mathrm{d}\Omega, \qquad \boldsymbol{F}(\boldsymbol{R}) = \int \boldsymbol{T}(\boldsymbol{R},\boldsymbol{s})\,\mathrm{d}\Omega. \tag{2.6),(2.7}$$

The integrations in eqs. (2.6) and (2.7) extend over the whole 4π-solid angle generated by the unit vector $\boldsymbol{s}$. If we define the spectral density $f(\boldsymbol{R},\boldsymbol{K})$ of the function $\Gamma(\boldsymbol{R},\boldsymbol{r})$ with respect to the variable $\boldsymbol{r}$ by the formula

$$f(\boldsymbol{R},\boldsymbol{K}) = (2\pi)^{-3}\int \Gamma(\boldsymbol{R},\boldsymbol{r})\exp(-\mathrm{i}\boldsymbol{K}\cdot\boldsymbol{r})\,\mathrm{d}^3K, \tag{2.8}$$

then, in view of eqs. (2.4)–(2.7), we obtain

$$Q(\boldsymbol{R},\boldsymbol{s}) = \frac{1}{c}\int_0^\infty f(\boldsymbol{R},\boldsymbol{K})\,K^2\,\mathrm{d}K, \tag{2.9}$$

$$\boldsymbol{T}(\boldsymbol{R},\boldsymbol{s}) = \frac{\boldsymbol{s}}{k_0}\int_0^\infty f(\boldsymbol{R},\boldsymbol{K})\,K^3\,\mathrm{d}K. \tag{2.10}$$

Equations (2.6) and (2.7) are analogous to the expressions of the phenomenological theory of radiative transfer for the "space density" of radiation $U(\boldsymbol{R})$ and for the net flux $\boldsymbol{F}(\boldsymbol{R})$, at $\boldsymbol{R}$, in terms of the specific intensity of radiation $I(\boldsymbol{R},\boldsymbol{s})$, viz.

$$U(\boldsymbol{R}) = \frac{1}{c}\int I(\boldsymbol{R},\boldsymbol{s})\,\mathrm{d}\Omega, \tag{2.11}$$

$$\boldsymbol{F}(\boldsymbol{R}) = \int \boldsymbol{s}\,I(\boldsymbol{R},\boldsymbol{s})\,\mathrm{d}\Omega. \tag{2.12}$$

A comparison of eq. (2.11) with eq. (2.6) suggests that the specific intensity of radiation $I(\boldsymbol{R},\boldsymbol{s})$ can be related to the angular component of the energy density $Q(\boldsymbol{R},\boldsymbol{s})$ by the simple equation

$$I(\boldsymbol{R},\boldsymbol{s}) = cQ(\boldsymbol{R},\boldsymbol{s}). \tag{2.13}$$

In view of eq. (2.9), we can write

$$I(\boldsymbol{R},\boldsymbol{s}) = \int_0^\infty f(\boldsymbol{R},\boldsymbol{K})\,K^2\,\mathrm{d}K. \tag{2.14}$$

The expression (2.8) for the spectral density $f(\boldsymbol{R},\boldsymbol{K})$, combined with eq. (2.14), gives a relationship between the specific intensity of radiation and the coherence function of the wavefield which seems to have been considered first by Ovchinnikov and Tatarskii [15]. Recently, on the basis of this definition, rigorous equations have been derived by the present author for radiative transfer of energy and momentum in free space in the presence of random source distribution [14].

It is clear from eqs. (2.7) and (2.10) that, with this definition of the specific intensity, the usual flux relation (2.7) is, in general, not obeyed. The condition under which the radiometric and field theoretic definitions of energy flux coincide, is given by

$$\int \boldsymbol{T}(\boldsymbol{R},\boldsymbol{s})\,\mathrm{d}\Omega = \int \boldsymbol{s}\,I(\boldsymbol{R},\boldsymbol{s})\,\mathrm{d}\Omega. \tag{2.15}$$

On substituting from eqs. (2.10) and (2.14) for $\boldsymbol{T}$ and I, eq. (2.15) becomes

$$\int \boldsymbol{s}(K - k_0)f(\boldsymbol{R},\boldsymbol{K})\,\mathrm{d}^3K = 0. \tag{2.16}$$

where, as before, $\boldsymbol{s}$ represents the unit vector along $\boldsymbol{K}$.

Consider a solution of eq. (2.16) of the form

$$f(\boldsymbol{R},\boldsymbol{K}) = I(\boldsymbol{R},\boldsymbol{s})\,\delta(K - k_0)/k_0^2. \tag{2.17}$$

On taking the Fourier inverse of eq. (2.8) and sub-

stituting for $f(\boldsymbol{R},\boldsymbol{K})$ from eq. (2.17), it follows that

$$\Gamma(\boldsymbol{R},\boldsymbol{r}) = \int I(\boldsymbol{R},\boldsymbol{s}) \exp(\mathrm{i}k_0 \boldsymbol{s}\cdot\boldsymbol{r})\, \mathrm{d}\Omega. \tag{2.18}$$

This representation of coherence function has been adopted by many authors in their investigations on the foundation of the theory of radiative energy transfer.

In a recent paper, Collett, Foley and Wolf [16] have shown that if, for propagation in free space, the coherence function $\Gamma(\boldsymbol{R},\boldsymbol{r})$ admits the representation eq. (2.18) then the wavefield $v(\boldsymbol{r})$ is necessarily statistically homogeneous. Furthermore one can establish the following theorem: *Theorem 2.1*: The coherence function $\Gamma(\boldsymbol{R},\boldsymbol{r})$ of the wavefield in a statistically homogeneous medium is independent of $\boldsymbol{R}$, (i.e., is a function of the difference variable $\boldsymbol{r}$ only) in the special case when the incident field, which satisfies the free space wave equation, is statistically homogeneous, at least, in the sense of second-order coherence theory.

It is also apparent from eqs. (2.8) and (2.14) that, when the coherence function of the wavefield $\Gamma(\boldsymbol{R},\boldsymbol{r})$ is a function of the difference variable $\boldsymbol{r}$ only, the specific intensity $I(\boldsymbol{R},\boldsymbol{s})$ is independent of position, i.e.,

$$I(\boldsymbol{R},\boldsymbol{s}) \equiv I(\boldsymbol{s}). \tag{2.19}$$

In that case the free space radiative energy transfer equation

$$\boldsymbol{s}\cdot\nabla_R I(\boldsymbol{R},\boldsymbol{s}) = 0 \tag{2.20}$$

is trivially satisfied.

Using these results, it may be shown that the representations eq. (2.17) or eq. (2.18) cannot be adopted to derive an equation of radiative transfer with non-vanishing extinction and scattering coefficients in statistically homogeneous media.

In this paper we consider wave propagation in a statistically *inhomogeneous* medium and show that the representation of eq. (2.17) for the spectral density $f(\boldsymbol{R},\boldsymbol{K})$ may be used when certain assumptions are made concerning the incident field and the randomly fluctuating medium.

3. Wave propagation in a weakly randomly fluctuating medium

The propagation of the space-dependent part $v(\boldsymbol{r})$ of the scalar wave in a medium with random fluctuations of the dielectric constant is governed by the stochastic scalar wave equation

$$(\nabla^2 + k_0^2 \epsilon(\boldsymbol{r}))v(\boldsymbol{r}) = 0, \tag{3.1}$$

where $k_0 = \omega/c$ is the wave number of the field in free space and $\epsilon(\boldsymbol{r})$ is the dielectric constant. If the fluctuating dielectric constant exhibits only small departures from its mean value, which for simplicity, is assumed to be unity, then a representation in terms of a parameter μ may be adopted [*1]:

$$\epsilon(\boldsymbol{r}) = 1 + \mu\epsilon_1(\boldsymbol{r}), \tag{3.2}$$

where

$$\mu \ll (k_0 l_1)^{-2}, \tag{3.3}$$

with l_1 denoting the smallest correlation distance of the dielectric constant fluctuation $\epsilon_1(\boldsymbol{r})$. Since we assumed $\langle\epsilon(\boldsymbol{r})\rangle = 1$, we have $\langle\epsilon_1(\boldsymbol{r})\rangle = 0$. Moreover the random function $\epsilon_1(\boldsymbol{r})$ is assumed to be a gaussian random variable.

On substituting from eq. (3.2) in eq. (3.1) we obtain

$$(\nabla^2 + k_0^2)v(\boldsymbol{r}) = -k_0^2 \mu\epsilon_1(\boldsymbol{r})v(\boldsymbol{r}). \tag{3.4}$$

If we recall the definition (eq. (2.1)) of the second-order coherence function of the wavefield $\Gamma(\boldsymbol{r}_1,\boldsymbol{r}_2)$, it follows from eq. (3.4) that $\Gamma(\boldsymbol{r}_1,\boldsymbol{r}_2)$ satisfies the equation

$$(\nabla_1^2 - \nabla_2^2)\Gamma(\boldsymbol{r}_1,\boldsymbol{r}_2) = -k_0^2 \mu[\langle\epsilon_1(\boldsymbol{r}_1)v(\boldsymbol{r}_1)v^*(\boldsymbol{r}_2)\rangle - \langle\epsilon_1(\boldsymbol{r}_2)v(\boldsymbol{r}_1)v^*(\boldsymbol{r}_2)\rangle]. \tag{3.5}$$

Let us assume that the field variable $v(\boldsymbol{r})$ can be expressed in a perturbation expansion [17,18]

$$v(\boldsymbol{r}) = v_0(\boldsymbol{r}) + \mu v_1(\boldsymbol{r}) + O(\mu^2). \tag{3.6}$$

Upon inserting from eq. (3.6) into eq. (3.4) and equating to zero the coefficients of the zeroth and of the first powers of μ, we obtain

$$(\nabla^2 + k_0^2)v_0(\boldsymbol{r}) = 0, \tag{3.7}$$

[*1] We assume max. $\epsilon_1(\boldsymbol{r}) \sim 1$. In that case μ represents the strength of the dielectric constant fluctuations.

$$(\nabla^2 + k_0^2)\, v_1(\boldsymbol{r}) = -k_0^2 \epsilon_1(\boldsymbol{r})\, v_0(\boldsymbol{r}). \tag{3.8}$$

The solution of eq. (3.8) can be expressed as

$$v_1(\boldsymbol{r}) = -k_0^2 \int G_0(\boldsymbol{r}-\boldsymbol{p})\, \epsilon_1(\boldsymbol{p})\, v_0(\boldsymbol{p})\, \mathrm{d}^3 p, \tag{3.9}$$

where

$$G_0(\boldsymbol{r}-\boldsymbol{p}) = -\frac{1}{4\pi} \frac{\exp(\mathrm{i}k_0|\boldsymbol{r}-\boldsymbol{p}|)}{|\boldsymbol{r}-\boldsymbol{p}|} \tag{3.10}$$

is the (outgoing) free space Green's function of the Helmholtz equation with wave number k_0.

We now expand the terms on the right hand side of eq. (3.5) to the lowest non-vanishing order in μ. Since $\langle \epsilon_1(\boldsymbol{r}) \rangle = 0$, it follows from that perturbation expansion (eq. (3.6)), (neglecting the terms of order μ^3), that

$$\begin{aligned}(\nabla_1^2 - \nabla_2^2)\, \bar{\Gamma}(\boldsymbol{r}_1,\boldsymbol{r}_2) &= k_0^4 \int [G_0(\boldsymbol{r}_1-\boldsymbol{p})\, \bar{\Gamma}_0(\boldsymbol{p},\boldsymbol{r}_2) \\ &+ G_0^*(\boldsymbol{r}_2-\boldsymbol{p})\, \bar{\Gamma}_0(\boldsymbol{r}_1,\boldsymbol{p})]\, \bar{B}(\boldsymbol{r}_1,\boldsymbol{p})\, \mathrm{d}^3 p \\ &- k_0^4 \int [G_0(\boldsymbol{r}_1-\boldsymbol{p})\, \bar{\Gamma}_0(\boldsymbol{p},\boldsymbol{r}_2) \\ &+ G_0^*(\boldsymbol{r}_2-\boldsymbol{p})\, \bar{\Gamma}_0(\boldsymbol{r}_1,\boldsymbol{p})]\, \bar{B}(\boldsymbol{p},\boldsymbol{r}_2)\, \mathrm{d}^3 p, \end{aligned} \tag{3.11}$$

where

$$\bar{B}(\boldsymbol{r}_1,\boldsymbol{r}_2) = B(\tfrac{1}{2}(\boldsymbol{r}_1+\boldsymbol{r}_2), \boldsymbol{r}_1-\boldsymbol{r}_2) = \mu^2 \langle \epsilon_1(\boldsymbol{r}_1)\, \epsilon_1(\boldsymbol{r}_2) \rangle, \tag{3.12}$$

$$\begin{aligned}\bar{\Gamma}_0(\boldsymbol{r}_1,\boldsymbol{r}_2) &= \Gamma_0(\tfrac{1}{2}(\boldsymbol{r}_1+\boldsymbol{r}_2), \boldsymbol{r}_1-\boldsymbol{r}_2) \\ &= \langle v_0(\boldsymbol{r}_1)\, v_0^*(\boldsymbol{r}_2) \rangle. \end{aligned} \tag{3.13}$$

We now change to the variables $\boldsymbol{R}$ and $\boldsymbol{r}$ which are defined by eq. (2.2). In terms of the new variables, eq. (3.11) can be shown after some rearrangements, to take the form

$$\begin{aligned}2\nabla_R \cdot \nabla_r \Gamma(\boldsymbol{R},\boldsymbol{r}) &= k_0^4 \int [G_0(\boldsymbol{p})\, B(\boldsymbol{R}+\tfrac{1}{2}\boldsymbol{r}-\tfrac{1}{2}\boldsymbol{p},\boldsymbol{p})\, \Gamma_0(\boldsymbol{R}-\tfrac{1}{2}\boldsymbol{p},\boldsymbol{r}-\boldsymbol{p}) \\ &- G_0^*(\boldsymbol{p})\, B(\boldsymbol{R}-\tfrac{1}{2}\boldsymbol{r}-\tfrac{1}{2}\boldsymbol{p},\boldsymbol{p})\, \Gamma_0(\boldsymbol{R}-\tfrac{1}{2}\boldsymbol{p},\boldsymbol{r}+\boldsymbol{p})]\, \mathrm{d}^3 p \\ &- k_0^4 \int [G_0(\boldsymbol{p})\, B(\boldsymbol{R}-\tfrac{1}{2}\boldsymbol{p},\boldsymbol{r}-\boldsymbol{p})\, \Gamma_0(\boldsymbol{R}-\tfrac{1}{2}\boldsymbol{p},\boldsymbol{r}-\boldsymbol{p}) \\ &- G_0^*(\boldsymbol{p})\, B(\boldsymbol{R}-\tfrac{1}{2}\boldsymbol{p},\boldsymbol{r}+\boldsymbol{p})\, \Gamma_0(\boldsymbol{R}-\tfrac{1}{2}\boldsymbol{p},\boldsymbol{r}+\boldsymbol{p})]\, \mathrm{d}^3 p. \end{aligned} \tag{3.14}$$

4. Derivation of the equation of radiative energy transfer

We will first make certain assumptions concerning the incident wavefield $v_0(\boldsymbol{r})$ and the dielectric constant fluctuation $\epsilon_1(\boldsymbol{r})$ which simplify eq. (3.14) considerably. We then proceed to derive the equation of radiative transfer satisfied by the specific intensity.

Let us assume that the incident field is statistically homogeneous, at least in the sense of the second-order correlation theory. The coherence function $\Gamma_0(\boldsymbol{R},\boldsymbol{r})$ then depends only on the difference variable $\boldsymbol{r}$, i.e.,

$$\Gamma_0(\boldsymbol{R},\boldsymbol{r}) \equiv \Gamma_0(\boldsymbol{r}). \tag{4.1}$$

Next we assume the random medium to be quasi-homogeneous, i.e., the coherence function $B(\boldsymbol{R},\boldsymbol{r})$ of the dielectric constant fluctuation is assumed to be a "slow" function of $\boldsymbol{R}$ and a "fast" function of $\boldsymbol{r}$. If L denotes the distance over which $B(\boldsymbol{R},\boldsymbol{r})$ remains sensibly constant with respect to its first argument, then, in view of the quasihomogeneity of the medium,

$$L \gg l_1, \tag{4.2a}$$

where l_1 is, as before, the smallest correlation distance of the fluctuations in the dielectric constant. Furthermore we assume L to be much larger than the correlation distance l_2 and the wavelength $2\pi/k_0$ of the incident field, i.e.,

$$L \gg l_2, \qquad L \gg 2\pi/k_0. \tag{4.2b,c}$$

These assumptions are satisfied in many problems of practical interest. A frequent model to describe a random medium e.g., the earth's atmosphere assumes quasihomogeneity. In many cases of optical propagation through atmosphere the conditions (eq. (4.2a)) and (eq. (4.2c)) are easily satisfied. The assumptions (eq. (4.1)) and (eq. (4.2b)) are obeyed by fields of thermal origin e.g., the blackbody radiation which is both statistically homogeneous and isotropic and whose correlation extends over distances of the order of the wavelength associated with the frequency of the radiation at which the energy transfer is being considered.

In view of the assumptions (eq. (4.1)) and (eq. (4.2)), we can simplify the various terms on the right hand side of eq. (3.14). Since the incident field is

considered statistically homogeneous, the first term can be written as

$$\int G_0(\boldsymbol{p})\, B(\boldsymbol{R}+\tfrac{1}{2}\boldsymbol{r}\quad \tfrac{1}{2}\boldsymbol{p},\boldsymbol{p})\, \Gamma_0(\boldsymbol{R}-\tfrac{1}{2}\boldsymbol{p},\boldsymbol{r}-\boldsymbol{p})\, \mathrm{d}^3p$$

$$= \int G_0(\boldsymbol{p})\, B(\boldsymbol{R}+\tfrac{1}{2}\boldsymbol{r}-\tfrac{1}{2}\boldsymbol{p},\boldsymbol{p})\, \Gamma_0(\boldsymbol{r}-\boldsymbol{p})\, \mathrm{d}^3p. \quad (4.3)$$

In view of our assumption (eq. (4.2b)), $B(\boldsymbol{R},\boldsymbol{r})$ is considered a "slow" function of $\boldsymbol{R}$ and $\Gamma_0(\boldsymbol{r})$ is considered a "fast" function of $\boldsymbol{r}$. Under these circumstances, we can replace $B(\boldsymbol{R}+\frac{1}{2}\boldsymbol{r}-\frac{1}{2}\boldsymbol{p},\boldsymbol{p})$ by $B(\boldsymbol{R},\boldsymbol{p})$ on the right hand side of eq. (4.3) without introducing an appreciable error. We then obtain

$$\int G_0(\boldsymbol{p})\, B(\boldsymbol{R}+\tfrac{1}{2}\boldsymbol{r}-\tfrac{1}{2}\boldsymbol{p},\boldsymbol{p})\, \Gamma_0(\boldsymbol{R}-\tfrac{1}{2}\boldsymbol{p},\boldsymbol{r}-\boldsymbol{p})\, \mathrm{d}^3p$$

$$\simeq \int G_0(\boldsymbol{p})\, B(\boldsymbol{R},\boldsymbol{p})\, \Gamma_0(\boldsymbol{r}-\boldsymbol{p})\, \mathrm{d}^3p. \quad (4.4)$$

Similarly,

$$\int G_0^*(\boldsymbol{p})\, B(\boldsymbol{R}-\tfrac{1}{2}\boldsymbol{r}-\tfrac{1}{2}\boldsymbol{p},\boldsymbol{p})\, \Gamma_0(\boldsymbol{R}-\tfrac{1}{2}\boldsymbol{p},\boldsymbol{r}+\boldsymbol{p})\, \mathrm{d}^3p$$

$$\simeq \int G_0^*(\boldsymbol{p})\, B(\boldsymbol{R},\boldsymbol{p})\, \Gamma_0(\boldsymbol{r}+\boldsymbol{p})\, \mathrm{d}^3p. \quad (4.5)$$

The third and fourth terms on the right hand side of eq. (3.14) can also be simplified by using eqs. (4.1) and (4.2b), and one obtains

$$\int G_0(\boldsymbol{p})\, B(\boldsymbol{R}-\tfrac{1}{2}\boldsymbol{p},\boldsymbol{r}-\boldsymbol{p})\, \Gamma_0(\boldsymbol{R}-\tfrac{1}{2}\boldsymbol{p},\boldsymbol{r}-\boldsymbol{p})\, \mathrm{d}^3p$$

$$\simeq \int G_0(\boldsymbol{p})\, B(\boldsymbol{R},\boldsymbol{r}-\boldsymbol{p})\, \Gamma_0(\boldsymbol{r}-\boldsymbol{p})\, \mathrm{d}^3p, \quad (4.6)$$

$$\int G_0^*(\boldsymbol{p})\, B(\boldsymbol{R}-\tfrac{1}{2}\boldsymbol{p},\boldsymbol{r}+\boldsymbol{p})\, \Gamma_0(\boldsymbol{R}-\tfrac{1}{2}\boldsymbol{p},\boldsymbol{r}+\boldsymbol{p})\, \mathrm{d}^3p$$

$$\simeq \int G_0^*(\boldsymbol{p})\, B(\boldsymbol{R},\boldsymbol{r}+\boldsymbol{p})\, \Gamma_0(\boldsymbol{r}+\boldsymbol{p})\, \mathrm{d}^3p. \quad (4.7)$$

On substituting from eqs. (4.4)–(4.7) in eq. (3.14) we obtain

$$2\nabla_R\cdot\nabla_r\Gamma(\boldsymbol{R},\boldsymbol{r}) = k_0^4\int [G_0(\boldsymbol{p})\,\Gamma_0(\boldsymbol{r}-\boldsymbol{p})$$

$$- G_0^*(\boldsymbol{p})\,\Gamma_0(\boldsymbol{r}+\boldsymbol{p})]\, B(\boldsymbol{R},\boldsymbol{p})\,\mathrm{d}^3p$$

$$- k_0^4\int [G_0(\boldsymbol{p})\, B(\boldsymbol{R},\boldsymbol{r}-\boldsymbol{p})\,\Gamma_0(\boldsymbol{r}-\boldsymbol{p})$$

$$- G_0^*(\boldsymbol{p})\, B(\boldsymbol{R},\boldsymbol{r}+\boldsymbol{p})\,\Gamma_0(\boldsymbol{r}+\boldsymbol{p})]\,\mathrm{d}^3p. \quad (4.8)$$

If, on the right-hand side, $\Gamma_0(\boldsymbol{r}+\boldsymbol{p})$ is replaced by $\Gamma(\boldsymbol{R},\boldsymbol{r}+\boldsymbol{p})$ then, in view of eq. (3.6), an error of the order μ^4 is introduced which is neglected in the present lowest non-vanishing order perturbative analysis [17]. Thus, to the second order in μ,

$$2\nabla_R\cdot\nabla_r\Gamma(\boldsymbol{R},\boldsymbol{r}) = k_0^4\int [G_0(\boldsymbol{p})\,\Gamma(\boldsymbol{R},\boldsymbol{r}-\boldsymbol{p})$$

$$G_0^*(\boldsymbol{p})\,\Gamma(\boldsymbol{R},\boldsymbol{r}+\boldsymbol{p})]\, B(\boldsymbol{R},\boldsymbol{p})\,\mathrm{d}^3p$$

$$- k_0^4\int [G_0(\boldsymbol{p})\, B(\boldsymbol{R},\boldsymbol{r}-\boldsymbol{p})\,\Gamma(\boldsymbol{R},\boldsymbol{r}-\boldsymbol{p})$$

$$- G_0^*(\boldsymbol{p})\, B(\boldsymbol{R},\boldsymbol{r}+\boldsymbol{p})\,\Gamma(\boldsymbol{R},\boldsymbol{r}+\boldsymbol{p})]\,\mathrm{d}^3p. \quad (4.9)$$

Next we take the Fourier inverse of eq. (2.8) to express $\Gamma(\boldsymbol{R},\boldsymbol{r})$ in terms of the spectral density $f(\boldsymbol{R},\boldsymbol{K})$, i.e.,

$$\Gamma(\boldsymbol{R},\boldsymbol{r}) = \int f(\boldsymbol{R},\boldsymbol{K})\exp(\mathrm{i}\boldsymbol{K}\cdot\boldsymbol{r})\,\mathrm{d}^3K. \quad (4.10)$$

Moreover we define the spectral density $\Phi(\boldsymbol{R},\boldsymbol{K})$ of the function $B(\boldsymbol{R},\boldsymbol{r})$ by means of Fourier decomposition with respect to $\boldsymbol{r}$.

$$B(\boldsymbol{R},\boldsymbol{r}) = \int \Phi(\boldsymbol{R},\boldsymbol{K})\exp(\mathrm{i}\boldsymbol{K}\cdot\boldsymbol{r})\,\mathrm{d}^3K. \quad (4.11)$$

It can be shown that $\Phi(\boldsymbol{R},\boldsymbol{K})$ is a real function. Inserting eq. (4.10) in eq. (4.9) and using eq. (4.11), we obtain after some rearrangement

$$\boldsymbol{K}\cdot\nabla_R f(\boldsymbol{R},\boldsymbol{K}) = -\tfrac{1}{2}\pi k_0^5\left|\int\Phi(\boldsymbol{R},\boldsymbol{K}-k_0\boldsymbol{s}')\,\mathrm{d}\Omega_{s'}\right| f(\boldsymbol{R},\boldsymbol{K})$$

$$+ \tfrac{1}{2}\pi k_0^3\,\delta(K-k_0)\int\Phi(\boldsymbol{R},\boldsymbol{K}-\boldsymbol{K}')\, f(\boldsymbol{R},\boldsymbol{K}')\,\mathrm{d}^3K'. \quad (4.12)$$

This integro-differential equation for $f(\boldsymbol{R},\boldsymbol{K})$ can be recast into the following integral equation [‡2]:

$$f(\boldsymbol{R},\boldsymbol{K}) = f_0(\boldsymbol{K})\exp\left\{-\int_0^\infty \mathrm{d}x''\, A(\boldsymbol{R}-x''\boldsymbol{s},\boldsymbol{K})\right\}$$

$$+ \delta(K-k_0)\int_0^\infty \mathrm{d}x'\, B(\boldsymbol{R}-x'\boldsymbol{s},\boldsymbol{K})$$

$$\times\exp\left\{-\int_0^{x'}\mathrm{d}x''\, A(\boldsymbol{R}-x''\boldsymbol{s},\boldsymbol{K})\right\}. \quad (4.13)$$

[‡2] The derivation of eq. (4.16) can be carried out on the same lines as in Ch. 2 (Sec. 6) of ref. [2].

where

$$f_0(\boldsymbol{K}) = \frac{1}{(2\pi)^3}\int \Gamma_0(\boldsymbol{r})\exp(-i\boldsymbol{K}\cdot\boldsymbol{r})\,\mathrm{d}^3r, \tag{4.14}$$

$$A(\boldsymbol{R},\boldsymbol{K}) = \frac{\pi k_0^5}{2K}\int \Phi(\boldsymbol{R},\boldsymbol{K}-k_0\boldsymbol{s}')\,\mathrm{d}\Omega_{s'}, \tag{4.15}$$

$$B(\boldsymbol{R},\boldsymbol{K}) = \frac{\pi k_0^3}{2K}\int \Phi(\boldsymbol{R},\boldsymbol{K}-\boldsymbol{K}')f(\boldsymbol{R},\boldsymbol{K}')\,\mathrm{d}K'. \tag{4.16}$$

Since the incident field is assumed to be statistically homogeneous, it can be shown that (ref. [12], appendix B)

$$f_0(\boldsymbol{K}) = I_0(\boldsymbol{s})\delta(K-k_0)/k_0^2. \tag{4.17}$$

On substituting eq. (4.17) in eq. (4.13), it becomes obvious that $f(\boldsymbol{R},\boldsymbol{K})$ is of the form

$$f(\boldsymbol{R},\boldsymbol{K}) = I(\boldsymbol{R},\boldsymbol{s})\delta(K-k_0)/k_0^2. \tag{4.18}$$

This relationship between the spectral density $f(\boldsymbol{R},\boldsymbol{K})$ and the specific intensity $I(\boldsymbol{R},\boldsymbol{s})$, which is a natural consequence of the assumptions made in this and the previous sections, has been shown, in section 2, to satisfy the "space density" relation (eq. (2.11)) and the energy flux relation (eq. (2.12)) of the phenomenological theory.

If this expression for $f(\boldsymbol{R},\boldsymbol{K})$ is now substituted in eq. (4.12), then after integration over $\boldsymbol{K}'$, $[\mathrm{d}^3K' = K'^2\mathrm{d}K'\,\mathrm{d}\Omega_{s'}]$, we obtain the following equation:

$$\boldsymbol{s}\cdot\nabla_R I(\boldsymbol{R},\boldsymbol{s}) = -\alpha(\boldsymbol{R},\boldsymbol{s})I(\boldsymbol{R},\boldsymbol{s}) + \int \beta(\boldsymbol{R},\boldsymbol{s},\boldsymbol{s}')I(\boldsymbol{R},\boldsymbol{s}')\,\mathrm{d}\Omega_{s'}, \tag{4.19}$$

where

$$\alpha(\boldsymbol{R},\boldsymbol{s}) = \tfrac{1}{2}\pi k_0^4\int \Phi(\boldsymbol{R},k_0(\boldsymbol{s}-\boldsymbol{s}'))\,\mathrm{d}\Omega_{s'}, \tag{4.20}$$

$$\beta(\boldsymbol{R},\boldsymbol{s},\boldsymbol{s}') = \tfrac{1}{2}\pi k_0^4\Phi(\boldsymbol{R},k_0(\boldsymbol{s}-\boldsymbol{s}')). \tag{4.21}$$

Eq. (4.19) is of the same form as the equation of radiative energy transfer of the phenomenological theory [2]. Our formulae (eq. (4.20)) and (eq. (4.21)) relate the extinction coefficient and the scattering coefficient β to the stochastic characteristics of the medium.

It is apparent from eqs. (4.20) and (4.21) that α and β are connected by the relation

$$\alpha(\boldsymbol{R},\boldsymbol{s}) = \int \beta(\boldsymbol{R},\boldsymbol{s},\boldsymbol{s}')\,\mathrm{d}\Omega_{s'}. \tag{4.22}$$

This equation, which is a consequence of the conservation of energy, implies that the extinction of a wave propagating along the direction $\boldsymbol{s}$ is due to the scattering in all other directions.

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PHOTON STATISTICS OF A TWO-PHOTON LASER ☆

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The steady state quantum statistical properties of a two-photon laser are presented for the case when the cavity loss mechanism is simulated by a single-photon process.

1. Introduction. The success of the quantum theory of lasers involving a single photon emission per atomic transition [1] has generated a great deal of interest in the possibility of achieving laser action involving the stimulated emission of two or more photons in a single atomic decay. The two-photon laser has the prospect of achieving high beam intensities since the strength of the coupling between the laser-active atoms and the light field is proportional to the light intensity in contrast to the square root of the light intensity in the one-photon laser.

Several authors [2–7] have studied the photon statistics of a two-photon laser on the basis of a model similar to the laser model of Scully and Lamb [1]. The model consists of a coupled system of a field and identical two-level atoms. The lasing levels of the atoms are assumed to have the same parity under the usual dipole approximation for the two-photon transition to take place. The atom–field interaction for the two-photon emission process may be described by the effective hamiltonian [8]:

$$H_I = g_2(\sigma^+ \hat{a}^2 + \sigma^- \hat{a}^{+2}), \tag{1}$$

where g_2 is the coupling constant given by

$$g_2 = \mu_{12}|E|^2/h^2 ,$$

μ_{12} is the matrix element for the two-photon transition, and $|E|^2$ has the dimension of field intensity, the atom operators σ and σ^+ are defined by

$$\sigma^+ = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}, \quad \sigma = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix},$$

and $\hat{a}, \hat{a}^+$ are the annihilation and creation operators of the field. The cavity losses due to the transmission of laser light through the end mirror are simulated by another set of two-level atoms which are pumped in the lower level at a constant rate and which can make a transition to the upper level by absorbing laser photons.

In most of the studies on the quantum statistical properties of the optical field of a two-photon laser [2,5,6], it is assumed that the end mirror of the cavity is transmitting at the sum frequency 2ω, i.e., the two-level atoms which simulate the cavity losses absorb two photons to make a transition from the lower level to the upper level. The restriction to a two-photon loss mechanism is made in order to retain the property of detailed balance. This model for the cavity losses is unrealistic since the photons do not escape in pairs. On the basis of this model, it is predicted that, in single- and two-mode two-photon lasers, the photon distribution functions are narrower than

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in the single-photon laser.

It is of considerable interest to investigate the quantum statistical properties of a two-photon laser when the cavity loss mechanism is simulated by a single-photon process. Some authors have approached this problem using perturbation methods [3,7]. In this paper, we present an exact solution of the master equation that describes a single-mode two-photon laser with a single-photon loss mechanism. We also derive expressions for the mean photon number and the relative fluctuations of the number of photons high above threshold.

2. *Photon distribution function.* We start with the equation of motion for the photon distribution function $p(n)$ in a single-mode two-photon laser. The master equation can be obtained in a straightforward manner by following the same method as that of the Scully–Lamb theory of a single-photon laser [1]. The resulting equation is

$$\frac{\partial p(n)}{\partial t} = -\frac{A_2(n+1)(n+2)}{1+(B_2/A_2)(n+1)(n+2)}p(n) + \frac{A_2 n(n-1)}{1+(B_2/A_2)n(n-1)}p(n-2) + C(n+1)p(n+1) - Cnp(n)\,, \tag{2}$$

where, similar to the Scully–Lamb theory, A_2 and B_2 are the gain and saturation parameters for the two-photon gain mechanism and C is the loss parameter which is related to the laser frequency ω and the Q of the cavity by the following equation:

$$C = \omega/Q\,. \tag{3}$$

In eq. (2), the term proportional to $p(n-2)$ represents the two-photon emission process, whereas the term proportional to $p(n+1)$ represents the single-photon absorption process. It is therefore evident that the detailed balance condition is not obeyed in the present case. This makes it more complicated to solve eq. (2) even in the steady state.

In an earlier paper [9] on the effect of cooperative atomic interactions on photon statistics in a single-mode laser, we encountered a master equation which also included two-photon emission and single-photon absorption processes. We solved this equation in steady state using a matrix approach. The same approach can be applied to the present problem.

In the steady state $[\partial p(n)/\partial t = 0]$, we obtain from eq. (2),

$$a_n p(n) + b_{n-2}p(n-2) + c_{n+1}p(n+1) = 0\,, \tag{4}$$

where

$$a_n = -\frac{(n+1)(n+2)}{1+(B_2/A_2)(n+1)(n+2)}\,, \quad b_n = \frac{(n+1)(n+2)}{1+(B_2/A_2)(n+1)(n+2)}\,, \quad c_n = (C/A_2)n\,. \tag{5a,b,c}$$

It is clear from eqs. (5a–c) that

$$a_n + b_n + c_n = 0\,. \tag{6}$$

In matrix notation, eq. (4) can be rewritten as

$$\begin{pmatrix} a_0 & c_1 & & & 0 \\ 0 & a_1 & c_2 & & \\ b_0 & 0 & \ddots & \ddots & \\ & b_1 & \ddots & & \\ 0 & & & & \end{pmatrix} \begin{pmatrix} p(0) \\ p(1) \\ \vdots \end{pmatrix} = 0\,. \tag{7}$$

It can be shown, by the method of induction, that the solution of eq. (7) is given by the following equation.

$$p(n) = \frac{(-1)^n p(0)}{c_1 \times \ldots \times c_n} M(n) , \tag{8}$$

where

$$M(n) = \det \begin{pmatrix} a_0 & c_1 & & & \\ 0 & a_1 & \ddots & 0 & \\ b_0 & 0 & \ddots & \ddots & \\ & \ddots & \ddots & \ddots & c_{n-1} \\ 0 & & b_{n-3} & 0 & a_{n-1} \end{pmatrix} \tag{9}$$

In eq. (8) $p(0)$ is determined from the normalization condition $\sum_{n=0}^{\infty} p(n) = 1$. By using the properties of the determinant and relation (6), the determinant $M(n)$ can be shown to be expressible in the following form:

$$M(n) = \det \begin{pmatrix} b_0 & c_1 & & 0 \\ & b_1 & c_1 & c_2 \\ & & b_2 & b_1 c_2 \\ 0 & & & b_1 + c_1 \end{pmatrix} \tag{10}$$

On substituting from eq. (10) in eq. (8), we obtain $p(n)$ as the following product of continued fractions:

$$p(n) = p(0) \prod_{r=1}^{n} \frac{1}{c_r} \; b_{r-1} + \cfrac{b_r c_{r-1}}{b_{r-2} + \cfrac{b_{r-1} c_{r-2}}{\ddots \; b_1 + \cfrac{b_0 c_1}{b_0}}} \tag{11}$$

This expression for $p(n)$ together with the expression of b_n and c_n [cf. eqs. (5b,c)] completely determines the photon statistics of a two-photon laser.

3. Photon number fluctuations high above threshold. We now consider the photon distribution function of the two-photon laser, when the laser operates high above threshold, a regime in which the photon distribution function of a single-photon laser is described by a Poisson distribution. For a Poisson distribution, the fluctuation in the number of photons is unity, i.e.,

$$\frac{(\Delta n)^2}{\langle n \rangle} = \frac{\langle n^2 \rangle - \langle n \rangle^2}{\langle n \rangle} = 1 . \tag{12}$$

For a two-photon laser, operating high above threshold, we have $\langle n \rangle = (A_2/B_2)^{1/2}$. Under this condition eq. (2) can be simplified considerably by neglecting 1 in the denominators of the first and second terms of the left hand side. We then obtain

$$\mathrm{d}p(n)/\mathrm{d}t = (A_2^2 B_2) p(n) + (A_2^2 B_2) p(n-2) - c(n+1) p(n+1) - Cnp(n) . \tag{13}$$

This equation can easily be solved, in steady state, using the following generation function:

$$g(x) = \sum_{n=0}^{\infty} x^n p(n) . \tag{14}$$

The generating function $g(x)$ obeys the following equation:

$$dg/dx - (A_2^2/B_2C)(1 + x)g = 0 . \tag{15}$$

This equation can be integrated and the resulting equation is

$$g(x) = \exp[(A_2^2/2B_2C)(x^2 + 2x - 3)] . \tag{16}$$

It is evident from eq. (14) that

$$\langle n \rangle = \left.\frac{dg}{dx}\right|_{x=1} , \quad \langle n^2 \rangle = \left(\frac{d^2g}{dx^2} + \frac{dg}{dx}\right)_{x=1} . \tag{17a, b}$$

It follows, on substituting from eq. (16) in eqs. (17a,b) that

$$\langle n \rangle = 2A_2^2/B_2C , \quad (\Delta n)^2/\langle n \rangle = \tfrac{3}{2} . \tag{18a,b}$$

According to eq. (18b), the photon distribution function for a two-photon laser is wider than the Poisson distribution. This result is in agreement with that of Golubev [7] that the relative fluctuations of the number of photons in a two-photon laser cannot be smaller than in coherent emission.

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Quantum statistics of multimode m-photon absorption process [a)]

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A density matrix method is used to obtain an exact solution for the reduced density matrix of the field in an arbitrary multimode m-photon absorption process. The results of some earlier specialized studies of photon statistics in multiphoton absorption process can be recovered from this solution.

It is well known that nonlinear interaction of light with matter changes the quantum statistical properties of the light field.[1] The changes of the photon statistics depend on the interaction process and on the initial conditions of the field.

In this paper we consider a multimode m-photon absorption process. The multiphoton absorption processes have recieved a great deal of attention in recent years due to the possibility of producing a radiation field which shows photon antibunching. The photon statistics of two-photon absorption process was studied using a generating function approach and exact expressions for the photon distribution function were given in single-mode[2-5] and two-mode[6] processes. The off-diagonal elements of the density matrix of the field in the single- and two-mode two-photon absorption processes were given by Simaan and Loudon.[7,8] Paul, Mohr, and Brunner[9] studied the photon statistics of m-photon absorption process on the basis of an approximate procedure. Recently, an exact analytic solution of the master equation describing single-mode m-photon absorption has been obtained by Zubairy and Yeh[10] using a density matrix approach and by Voigt, Bandilla, and Ritze[11] using a Laplace transform method.

In the present paper we extend these results to arbitrary multimode absorption process. We begin by considering the master equation which describes a multimode m-photon process. We present the exact solution of the reduced density matrix of the field using the matrix approach.[10] Due to the general nature of our problem the results of the earlier studies can be recovered in the appropriate limits.

We consider a coupled system of a field and N noninteracting two-level atoms in their ground state. The number of atoms N in the lower level are assumed to be maintained constant by some external influence. We assume that the atoms make a transition from the lower level to the upper level by absorbing m photons; x_i photons in mode i ($i = 1, 2, \ldots, l$; $l \leq m$). We then obtain

$$\sum_{i=1}^{l} x_i = m. \tag{1}$$

The single-mode situation corresponds to the case when one of the x_i's such as x_1 is equal to m and all the rest are zero. The reduced density matrix $\hat{\rho}_f$ of the field can then be shown to satisfy the following equation of motion using the standard perturbation techniques

$$\frac{d\hat{\rho}_f}{dt} = -\beta^{(m)}\left[\left(\prod_{i=1}^{l}\hat{a}_i^{\dagger x_i}\right)\left(\prod_{i=1}^{l}\hat{a}_i^{x_i}\right)\hat{\rho}_f - 2\left(\prod_{i=1}^{l}\hat{a}_i^{x_i}\right)\hat{\rho}_f \times\left(\prod_{i=1}^{l}\hat{a}_i^{\dagger x_i}\right) + \hat{\rho}_f\left(\prod_{i=1}^{l}\hat{a}_i^{\dagger x_i}\right)\left(\prod_{i=1}^{l}\hat{a}_i^{x_i}\right)\right], \tag{2}$$

where $\beta^{(m)}$ is the absorption coefficient for m-photon absorption, and $\hat{a}^\dagger$, $\hat{a}$ are the photon creation and destruction operators of the ith mode, respectively. In Eq. (2) the saturation of the absorbing atoms is neglected.

We denote the state in which there are n_i photons in the ith mode ($i = 1, 2, \ldots, l$) by $|n_1, n_2 \ldots n_l\rangle$. The equation of motion (2) for $\hat{\rho}_f$ can be translated into an equation for the matrix element

$$\rho(n_1, \ldots, n_l; n_1 + K_1, \ldots, n_l + K_l, \tau) = \langle n_1, \ldots, n_l|\hat{\rho}_f|n_1 + K_1, \ldots, n_l + K_l\rangle, \tag{3}$$

of the reduced density matrix by evaluating the matrix element of each term in Eq. (2) between the appropriate Fock states. The resulting equation is

$$\frac{\partial\rho(n_1, \ldots, n_l; n_1 + K_1, \ldots, n_l + K_l;\tau)}{\partial\tau} = a(n_1, \ldots, n_l; n_1 + K_1, \ldots, n_l + K_l)\rho(n_1, \ldots, n_l; n_1 + K_1, \ldots, n_l + K_l; \tau) + b(n_1 + x_1, \ldots, n_l + x_l; n_1 + x_1 + K_1, \ldots, n_l + x_l + K_l) \times\rho(n_1 + x_1, \ldots, n_l + x_l; n_1 + x_1 + K_1, \ldots, n_l + x_l + K_l; \tau), \tag{4}$$

where $\tau = 2\beta^{(m)}t$ and

$$a(n_1, \ldots, n_l; n_1', \ldots, n_l') = -\frac{1}{2}\left[\prod_{i=1}^{l}\frac{n_i!}{(n_i - x_i)!} + \prod_{i=1}^{l}\frac{n_i'!}{(n_i' - x_i)!}\right], \tag{5a}$$

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$$b(n_1, \ldots, n_l; n_1', \ldots, n_l') = \left[\prod_1^l \frac{n_j! n_j'!}{(n_j - x_j)!(n_j' - x_j)!}\right]^{1/2}. \tag{5b}$$

Following the method employed in Ref. 10, we first express Eq. (4) in the matrix notation

$$\frac{\partial}{\partial \tau}\rho_{i_1,\ldots,i_l}(\tau) = M_{i_1,\ldots,i_l}\rho_{i_1,\ldots,i_l}(\tau), \tag{6}$$

where

$$\rho_{i_1,\ldots,i_l}(\tau) = \begin{bmatrix} \rho(i_1, \ldots, i_l;\ i_1 + K_1, \ldots, i_l + K_l;\ \tau) \\ \rho(i_1 + x_1, \ldots, i_l + x_l;\ i_1 + K_1 + x_1, \ldots, i_l + K_l + x_l;\ \tau) \\ \vdots \\ \rho(i_1 + nx_1, \ldots, i_l + nx_l;\ i_1 + K_1 + nx_1, \ldots i_l + K_l + nx_l;\ \tau) \\ \vdots \end{bmatrix}, \tag{7}$$

$$M_{i_1,\ldots,i_l} = \begin{bmatrix} m_{00} & m_{01} & & & \\ & m_{11} & m_{12} & \bigcirc & \\ & \bigcirc & & & \\ & & & m_{nn} & m_{n,n+1} \end{bmatrix}. \tag{8}$$

In Eq. (8), the elements m_{jk} are given by the following expressions

$$m_{jj} = a(i_1 + jx_1, \ldots, i_l + jx_l;\ i_1 + K_1 + jx_1, \ldots, i_l + K_l + jx_l), \tag{9a}$$

$$m_{j,j+1} = b(i_1 + (j+1)x_1, \ldots, i_l + (j+1)x_l;\ i_1 + K_1 + (j+1)x_1, \ldots, i_l + K_l + (j+1)x_l), \tag{9b}$$

for $j = 0, 1, 2, \cdots$.

The solution of Eq. (6) or Eq. (4) can be expressed in the form

$$\rho(i_1 + nx_1, \ldots, i_l + nx_l;\ i_1 + K_1 + nx_1, \ldots, i_l + K_l + nx_l;\tau) = \sum_{s=0}^{n}\sum_{q=0}^{n} \alpha_n^{i_1,\ldots,i_l}\beta_q^{i_1,\ldots,i_l} e^{\lambda_{i_1,\ldots,i_l}\tau}\rho(i_1 + qx_1, \ldots, i_l + qx_l; \times i_1 + K_1 + qx_1, \ldots, i_l + K_l + qx_l;\ 0), \tag{10}$$

where $\lambda_{i_1,\ldots,i_l}$ are the eigenvalues of the matrix $M_{i_1,\ldots,i_l}$, i.e., they satisfy the equation,

$$\det[M_{i_1,\ldots,i_l} - \lambda I] = 0, \tag{11}$$

(I being the unit matrix), $\alpha_n^{i_1,\ldots,i_l}$ is the nth element of the right eigenstate of $M_{i_1,\ldots,i_l}$ corresponding to the eigenvalue $\lambda_{i_1,\ldots,i_l}$, and $\beta_q^{i_1,\ldots,i_l}$ is the qth element of the left eigenstate of M corresponding to the eigenvalue $\lambda_{i_1,\ldots,i_l}$. The matrix elements $\alpha_n^{i_1,\ldots,i_l}$ and $\beta_q^{i_1,\ldots,i_l}$ can be shown to obey the following recursion relations

$$m_{n,n+1}\alpha_n^{i_1,\ldots,i_l} + m_{n,n}\alpha_n^{i_1,\ldots,i_l} = m_{jj}\alpha_n^{i_1,\ldots,i_l}, \tag{12a}$$

$$m_{q-1,q}\beta_q^{i_1,\ldots,i_l} + m_{qq}\beta_q^{i_1,\ldots,i_l} = m_{jj}\beta_q^{i_1,\ldots,i_l}. \tag{12b}$$

By solving Eq. (11) and by iterating the recursion relations (12a) and (12b), we obtain

$$\lambda_{i_1,\ldots,i_l} = m_{ss}, \tag{13}$$

$$\alpha_n^{i_1,\ldots,i_l} = \begin{cases} \prod_{r=s}^{n-1}\left(\frac{m_{r,r+1}}{m_{ss} - m_{r+1,r+1}}\right); & n > s, \\ 0; & n < s, \end{cases} \tag{14}$$

$$\beta_q^{i_1,\ldots,i_l} = \begin{cases} \prod_{r=s}^{q-1}\left(\frac{m_{r,r+1}}{m_{ss} - m_{rr}}\right); & q > s, \\ 0; & q < s. \end{cases} \tag{15}$$

If we let $n = 0$, $i_j = n_j$, and $K_j = n_j' - n_j$ $(j = 1, 2 \ldots l)$ in Eq. (10), then, on substituting from Eqs. (9a), (9b), and (13)–(15), it follows

$$\rho(n_1, \ldots, n_l; n_1', \ldots n_l';\ \tau) = \sum_{s=0}^{r}\sum_{q=s}^{r} \frac{\Pi_{r=s}^{q-1} b(n_1 + rx_1, \ldots, n_l + rx_l; n_1' + rx_1, \ldots, n_l' + rx_l)}{\Pi_{r\neq s}^{q} [a(n_1 + sx_1, \ldots, n_l + sx_l; n_1' + sx_1, \ldots, n_l' + sx_l) - a(n_1 + rx_1, \ldots, n_l + rx_l;\ n_1' + rx_1, \ldots, n_l' + rx_l)]} \times e^{a(n_1 + sx_1, \ldots, n_l + sx_l; n_1' + sx_1, \ldots, n_l' + sx_l)\tau}\rho(n_1 + qx_1, \ldots, n_l + qx_l; n_1' + qx_1, \ldots n_l' + qx_l, 0). \tag{16}$$

This equation, combined with the expressions for a's and b's [cf. Eqs. (5a) and (5b)], completely determines the time evolution of the density matrix. The photon distribution function $p(n_1, \ldots, n_l;\ \tau) = \langle n_1, \ldots, n_l|\hat{\rho}_t|n_1, \ldots, n_l\rangle$ can be determined from Eq. (16) by putting $n_j' = n_j$ $(j = 1, \ldots, l)$.

We now show how some of the earlier specialized results regarding multiphoton absorption processes can be obtained from Eq. (16).

In the case of single-mode m-photon absorption process, one of the x_j's (say x_1) is equal to m and all the rest are

zero. We then obtain, from Eqs. (5a) and (5b), that

$$a(n_1;\ n_1') = -\frac{1}{2}\left(\frac{n_1!}{(n_1-m)!} + \frac{n_1'!}{(n_1'-m)!}\right), \tag{17a}$$

$$b(n_1;\ n_1') = \left(\frac{n_1!n_1'!}{(n_1-m)!(n_1'-m)!}\right)^{1/2}. \tag{17b}$$

If follows from Eq. (16) that

$$\rho(n_1;\ n_1';\ \tau) = \sum_{s=0}^{\infty}\sum_{q}\frac{\Pi_{r=1}^{q}\, b(n_1+mr;\ n_1'+mr)}{\Pi_{r=1}^{q}[a(n_1+ms;\ n_1'+ms)-a(n_1+mr;\ n_1'+mr)]}\, e^{a(n_1+ms,n_1'+ms)\tau}\rho(n_1+mq;\ n_1'+mq;\ 0). \tag{18}$$

After making some rearrangements, this equation can be shown to be identical to Eq. (31) in Ref. 10 (see also Ref. 11 for the case $n_1 = n_1'$). We have discussed some aspects of photon statistics in single-mode m-photon abosorption process, such as photon antibunching, in that paper.

As another example, we consider m-mode m-photon absorption process. In this case, $l = m$ and $x_i = 1 (i = 1, 2, ..., m)$. The expressions for a and b in Eqs. (5a) and (5b) then become

$$a(n_1, ..., n_m;\ n_1', ..., n_m') = -\tfrac{1}{2}[n_1 ..., n_m + n_1' ..., n_m'], \tag{19a}$$

$$b(n_1, ..., n_m;\ n_1', ..., n_m') = (n_1 ..., n_m n_1' ..., n_m')^{1/2}. \tag{19b}$$

Moreover, from Eq. (16), we obtain the following solution for the density matrix

$$\rho(n_1, ..., n_m;\ n_1', ..., n_m';\ \tau) = \sum_{s=0}^{\infty}\sum_{q}\frac{\Pi_{r=1}^{q}\, b(n_1+r, ..., n_m+r;\ n_1'+r, ...n_m+r)}{\Pi_{r=1}^{q}[a(n_1+s_1, ..., n_m+s;\ n_1'+s, ..., n_m'+s)-a(n_1+r, ..., n_m+r;\ n_1'+r, ..., n_m'+r]}$$

$$\times e^{a(n_1+\cdots n_m+\cdots n_1'+\cdots n_m'+\cdots)\tau}\rho(n_1+q, ..., n_m+q;\ n_1'+q, ..., n_m'+q;\ 0). \tag{20}$$

Simaan and Loudon[6-8] have discussed the case $m = 2$ in some detail. Equation (20) together with Eqs. (19a) and (19b), after some rearrangement, can be shown to reduce to their results. For a discussion of the quantum statistical properties of the double-beam two-photon absorption process, we refer the reader to Ref. 6.

In conclusion, we have obtained an exact solution of the master equation that describes an arbitrary multimode multiphoton process. We have shown via two examples how the results of the earlier specialized studies can be recovered from this general solution.

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Intensity correlations in a two-mode laser with coupled transitions

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Intensity correlations in a two-mode laser oscillating on two coupled transitions have been studied by means of the Fokker-Planck equation. It is shown that they can be expressed in terms of the eigenvalues and eigenfunctions of a one-dimensional Schrodinger-type equation. It is found that near threshold intensity correlations cannot be approximated by a single exponential. For moderately large excitations, however, a single exponential dominates. Approximate analytic expressions for large excitations have been obtained which show that the correlation time increases with increasing excitation.

I. INTRODUCTION

Recently a quantum-mechanical treatment of laser oscillations on two coupled atomic transitions has been given.[1] In that treatment a set of homogeneously broadened three-level atoms interacting with a two-mode electromagnetic field in a unidirectional-ring configuration was considered. The atomic-level configuration that was considered is usually referred to as the "Λ" configuration in the literature on three-level atomic media.[2] It was assumed that each transition supported a distinct mode of the electromagnetic field. A master equation for the photon distribution function was derived under the conditions of perfect resonance between the atomic transition and the corresponding mode frequencies. This equation was solved in the steady state and the fluctuation properties of the optical field were discussed. One of the principal conclusions was that the intensity fluctuations do not, in general, die away with increasing excitation. Small asymmetries in gain may change the statistical properties of the two modes significantly. These effects are, of course, the result of mode competition. The question that presents itself now is how does the mode competition affect the correlations and other time-dependent phenomena. This paper answers this question partly.

II. TIME-DEPENDENT FOKKER-PLANCK EQUATION

The starting point of our discussion is the master equation for the photon distribution function $p(n_1, n_2)$ derived in Ref. 1, viz.,

$$\begin{aligned}\frac{d}{dt}p(n_1,n_2) = &-\frac{A(n_1+1)}{1+(B/A)(n_1+n_2+2)}p(n_1,n_2)-\frac{A(n_2+1)}{1+(B/A)(n_1+n_2+2)}p(n_1,n_2)\\&+C_1(n_1+1)p(n_1+1,n_2)+C_2(n_2+1)p(n_1,n_2+1)\\&+\frac{An_1}{1+(B/A)(n_1+n_2+1)}p(n_1-1,n_2)+\frac{An_2}{1+(B/A)(n_1+n_2+1)}p(n_1,n_2-1)\\&-C_1n_1p(n_1,n_2)-C_2n_2p(n_1,n_2)\,.\end{aligned}\tag{1}$$

Here $p(n_1, n_2) = \rho(n_1 n_2; n_1 n_2; t)$ is the diagonal element of the density matrix operator of the optical field; n_1, n_2 are the occupation numbers of the two modes and A, B, and C_i are the gain, saturation, and the loss coefficients, respectively.[1] This equation will now be converted into an equation for the quasiprobability distribution function Φ for the complex field amplitudes by using the coherent-state representation of the density matrix of the electromagnetic field.[3] The Fock-state elements of the density matrix operator are related to Φ by

$$\rho(n_1n_2;n_1'n_2';t) = \int \Phi(\beta_1,\beta_1^*,\beta_2,\beta_2^*,t)\frac{e^{-|\beta_1|^2-|\beta_2|^2}\beta_1^{n_1}\beta_1^{*n_1'}\beta_2^{n_2}\beta_2^{*n_2'}}{(n_1!\,n_1'!\,n_2!\,n_2'!)^{1/2}}d^2\beta_1 d^2\beta_2\,,\tag{2}$$

where β_i is the complex field amplitude associated with the ith mode in the coherent state of the field. In what follows, our interest will be confined mainly to the intensity correlations so that we will not be concerned with the dependence of Φ on the phases β_1 and β_2. Such dependence would be needed for a discussion of the amplitude correlations but the problem in that case turns out to be rather complex. Now, the relevant intensity distribution function $P(I_1, I_2, t)$ that is needed to calculate intensity correlations can be projected out of Φ simply by noting that it is determined solely by the diagonal Fock-state matrix elements of the density operator. The two are related by

$$\rho(n_1 n_2; n_1 n_2; t) = p(n_1, n_2, t) = \int P(I_1, I_2, t) e^{-I_1 - I_2} \frac{I_1^{n_1}}{n_1!} \frac{I_2^{n_2}}{n_2!} dI_1 dI_2 . \tag{3}$$

We also introduce an auxiliary function by

$$w(n_1, n_2) = \left(1 + \frac{B}{A}(n_1 + n_2 + 2)\right)^{-1} p(n_1, n_2) = \int \bar{w}(I_1, I_2, t) e^{-I_1 - I_2} \frac{I_1^{n_1}}{n_1!} \frac{I_2^{n_2}}{n_2!} dI_1 dI_2 . \tag{4}$$

Using Eq. (4) we obtain the following set of coupled differential equations:

$$\frac{\partial}{\partial t} P(I_1, I_2, t) = -A\left(\frac{\partial}{\partial I_1} I_1 + \frac{\partial}{\partial I_2} I_2 - \frac{\partial}{\partial I_1} I_1 \frac{\partial}{\partial I_1} - \frac{\partial}{\partial I_2} I_2 \frac{\partial}{\partial I_2}\right) \bar{w}(I_1, I_2, t) + \left(C_1 \frac{\partial}{\partial I_1} I_1 + C_2 \frac{\partial}{\partial I_2} I_2\right) p(I_1, I_2, t) \tag{5a}$$

and

$$\left[1 - \frac{B}{A}\left(I_1 \frac{\partial}{\partial I_1} + I_2 \frac{\partial}{\partial I_2} - I_1 - I_2\right)\right] \bar{w}(I_1, I_2, t) = P(I_1, I_2, t) . \tag{5b}$$

These equations can be solved exactly in the steady state and the corresponding solution was discussed in Ref. 1. The general time-dependent problem for $p(I_1, I_2, t)$ is very complicated because it does not seem feasible to obtain a closed equation either for $\bar{w}$ or p. However, if we take recourse to the scaling argument a closed equation for P can be obtained. This argument works as follows: since B/A is typically $\sim 10^{-6}$ so that to a very good approximation the derivative terms $(B/A) I_i (\partial / \partial I_i)$ can be ignored compared to $(B/A) I_i$ in Eq. (5b) which then is easily solved to give

$$\bar{w} = \frac{P}{1 + (B/A)(I_1 + I_2)} . \tag{6}$$

On substituting Eq. (6) into Eq. (5a) we obtain the following closed equation for $P(I_1, I_2, t)$:

$$\frac{dP}{dt} = -A\left(\frac{\partial}{\partial I_1} I_1 + \frac{\partial}{\partial I_2} I_2 - \frac{\partial}{\partial I_1} I_1 \frac{\partial}{\partial I_1} - \frac{\partial}{\partial I_2} I_2 \frac{\partial}{\partial I_2}\right)\left(1 + \frac{B}{A}(I_1 + I_2)\right)^{-1} P + \left(C_1 \frac{\partial}{\partial I_1} I_1 + C_2 \frac{\partial}{\partial I_2} I_2\right) P . \tag{7}$$

Note that Eq. (7) has the formal structure of a Fokker-Planck equation and is the desired equation that will form the basis of the discussion in the rest of the paper. We emphasize that the approximation made to reach at Eq. (7) is not the weak-signal-limit approximation and does not restrict the validity of the above approach to the region near threshold.

The effects of mode competition are more interesting when both the modes have equal losses. We shall therefore consider this case first and put $C_1 = C_2 = C$ in Eq. (7). The general problem with unequal losses is still quite complicated. We shall consider this problem later. With equal losses for the modes we now look for the general solution $P(I_1, I_2, t)$ of the form

$$P(I_1, I_2, t) = \sum_{Lm} C_{Lm} f_{Lm}(I_1, I_2) e^{-\lambda_{Lm} t} , \tag{8}$$

where C_{Lm} are the coefficients to be determined by the boundary conditions and λ_{Lm} is the eigenvalue of the differential operator on the right-hand side of Eq. (7) associated with the eigenfunction f_{Lm}. The differential operator depends on two variables I_1 and I_2, therefore the eigenvalues and the eigenfunctions are labeled by two integers L, m. The eigenvalue equation is obtained from Eqs. (7) and (8) to be

$$-A\left(\frac{\partial}{\partial I_1} I_1 + \frac{\partial}{\partial I_2} I_2 - \frac{\partial}{\partial I_1} I_1 \frac{\partial}{\partial I_1} - \frac{\partial}{\partial I_2} I_2 \frac{\partial}{\partial I_2}\right)\left(1 + \frac{B}{A}(I_1 + I_2)\right)^{-1} f_{Lm} + C\left(\frac{\partial}{\partial I_1} I_1 + \frac{\partial}{\partial I_2} I_2\right) f_{Lm} = -\lambda_{Lm} f_{Lm} . \tag{9}$$

In order to solve Eq. (9) we first try a substitution of the form

$$f_{Lm}(I_1, I_2) = N^{-1} e^{-U(I_1, I_2)} g_{Lm}(I_1, I_2) , \tag{10}$$

where $U(I_1, I_2)$ is some function of I_1 and I_2 which will be chosen to yield a self-adjoint equation for g_{Lm} and N is a normalization constant. It is easy to show that the choice

$$U(I_1, I_2) = -\ln\left(1 + \frac{B}{A}(I_1 + I_2)\right) - \frac{A - C}{2A}(I_1 + I_2) + \frac{BC}{4A^2}(I_1 + I_2)^2 \tag{11}$$

yields the following self-adjoint equation for g_{Lm}:

$$4\left\{\frac{\partial}{\partial I_1} I_1 \frac{\partial}{\partial I_1} + \frac{\partial}{\partial I_2} I_2 \frac{\partial}{\partial I_2} - \left[\frac{B^2C^2}{4A^4}(I_1 + I_2)^4 - \frac{BC(A - C)}{2A^3}(I_1 + I_2)^3 + \left(\frac{(A - C)^2}{4A^2} - \frac{3}{2}\frac{BC}{A^2}\right)(I_1 + I_2)^2 + \frac{A - C}{A} - \left(1 + \frac{B}{A}(I_1 + I_2)\right)\frac{\lambda_{Lm}}{A}\right]\right\} g_{Lm} = 0 . \tag{12}$$

Notice that the steady-state solution corresponds to $L = 0 = m$, $\lambda_{Lm} = 0$ and is found from Eq. (9) after straightforward integration to be

$$p_s(I_1, I_2) = Q^{-1}\left(1 + \frac{B}{A}(I_1 + I_2)\right) \times \exp\left(\frac{A - C}{A}(I_1 + I_2) - \frac{BC}{2A^2}(I_1 + I_2)^2\right) . \tag{13}$$

If we choose $N = \sqrt{Q}$, the relation [Eq. (10)] can be rewritten for later convenience in the form

$$f_{Lm}(I_1, I_2) = \sqrt{p_s}\,[1 + (B/A)(I_1 + I_2)]^{1/2} g_{Lm}(I_1, I_2) . \tag{14}$$

The functions g_{Lm} may be chosen to be an orthonormal set. In addition we assume that they form a complete set. The two conditions can be expressed in terms of the eigenfunctions f_{Lm} as

$$\int \frac{f^*_{L'm'}(I_1, I_2) f_{Lm}(I_1, I_2)}{p_s(I_1, I_2)} dI_1 dI_2 = \delta_{LL'}\delta_{mm'} \tag{15}$$

and

$$\sum_{Lm} \frac{f^*_{Lm}(I_1, I_2) f_{Lm}(I_1', I_2')}{p_s(I_1', I_2')} = \delta(I_1 - I_1')(I_2 - I_2') . \tag{16}$$

To calculate two-time correlations we shall need the Green's function $G(I_1, I_2, t \mid I_1^0, I_2^0, t_0)$ which is also the conditional probability for the intensities to be characterized by the values I_1 and I_2 at time t given their values I_1^0, I_2^0 at t_0. To this end we note that $G(I_1, I_2, t \mid I_1^0, I_2^0, t_0)$ is the solution $P(I_1, I_2, t)$ of the Fokker-Planck equation (7) with the initial condition $P(I_1, I_2, t_0) = \delta(I_1 - I_1^0)\delta(I_2 - I_2^0)$. It follows immediately that

$$G(I_1, I_2, t \mid I_1^0, I_2^0, t_0) = \sum_{Lm} \frac{f^*_{Lm}(I_1, I_2) f_{Lm}(I_1^0, I_2^0)}{p_s(I_1^0, I_2^0)} e^{-\lambda_{Lm}(t - t_0)}, \quad t > t_0 . \tag{17}$$

The two-time joint probability density $p_2(I_1, I_2, t; I_1', I_2', t')$ for the intensities at two different times can be obtained easily now. Noting that the stationary-state probabilities are independent of the origin of time we obtain

$$p_2(I_1, I_2, t + T; I_1', I_2', t) = G(I_1, I_2, T \mid I_1', I_2', 0) p_s(I_1', I_2') = \sum_{Lm} f^*_{Lm}(I_1, I_2) f_{Lm}(I_1', I_2') e^{-\lambda_{Lm} T}, \quad T \geq 0 . \tag{18}$$

For discussing second-order intensity correlations, we need the two-time joint probability density, which we have been able to express in terms of certain eigenvalues and eigenfunctions. For discussing higher-order intensity correlations, higher-order probabilities will be required, which also can be expressed in terms of the same set of eigenfunctions and eigenvalues. Our problem now is to solve for the eigenvalues and the eigenfunctions.

III. SOLUTIONS OF THE EIGENVALUE PROBLEM

The eigenvalue equation (12) can be written in a simpler form in terms of two new variables defined by

$$u = I_1 + I_2, \quad u > 0 \tag{19a}$$

$$v = \frac{(I_1 - I_2)}{(I_1 + I_2)}, \quad -1 \leq v \leq 1 \tag{19b}$$

or

$$I_j = \frac{u}{2}[1 - (-1)^j v] . \tag{19c}$$

We also have

$$dI_1 dI_2 = \frac{u}{2} du\, dv . \tag{20}$$

Then g_{Lm} obeys the following equation:

$$\left[\frac{\partial}{\partial u} u^2 \frac{\partial}{\partial u} - \left(\frac{B^2C^2}{4A^2}u^4 - \frac{BC(A - C)}{2A^3}u^3\right) + \left(\frac{(A - C)^2}{4A^2} - \frac{3}{2}\frac{BC}{A^2}\right)u^2 + u\frac{(A - C)}{A} - u\left(1 + \frac{B}{A}u\right)\frac{\lambda_{Lm}}{A} + \frac{\partial}{\partial v}(1 - v^2)\frac{\partial}{\partial v}\right] g_{Lm} = 0 . \tag{21}$$

The form of Eq. (21) suggests a solution of the form

$$g_{Lm}(u, v) = R_{Lm}(u) S_L(v) . \tag{22}$$

Substitution of this into Eq. (21) yields the following two uncoupled equations for $R_{Lm}(u)$ and $S_L(v)$:

$$\frac{d}{du} u^2 \frac{d}{du} R_{Lm} - \left[\frac{B^2C^2}{4A^2}u^4 - \frac{BC(A - C)}{2A^3}u^3 + \left(\frac{(A - C)^2}{4A^2} - \frac{3}{2}\frac{BC}{A^2}\right)u^2 + u\left(\frac{A - C}{A}\right) - u\left(1 + \frac{B}{A}u\right)\frac{\lambda_{Lm}}{A} + \rho_L\right] R_{Lm} = 0 \tag{23a}$$

and

$$\frac{d}{dv}(1-v^2)\frac{d}{dv}S_L+\beta_L S_L=0, \tag{23b}$$

where β_L is the separation constant. With the choice $\beta_L=(L-4)(L+2)=l(l+1)$, $l=0,1,2,$ Eq. (23b) is just the Legendre equation and S_L can be expressed in terms of P_l, the Legendre polynomial of order l as

$$S_L(v)=\left(\frac{2l+1}{2}\right)^{1/2}P_l(v). \tag{24}$$

The eigenvalue equation (23) can be reduced to an even simpler form by making a change of variable

$$u=r^2,\quad \frac{u}{2}du=r^4dr,$$
$$R_{Lm}(r^2)=r^{-3/2}\psi_{Lm}(r). \tag{25}$$

The equation satisfied by ψ_{Lm} is

$$\frac{d^2}{dr^2}\psi_{Lm}-\left\{\frac{B^2C^2}{A^4}r^6-\frac{2BC(A-C)}{A^3}r^4+\left[\left(\frac{A-C}{A}\right)^2-\frac{6BC}{A^2}\right]r^2+\frac{4(A-C)}{A}-4\left(1+\frac{B}{A}r^2\right)\frac{\lambda_{Lm}}{A}+\frac{L(L+2)+1}{r^2}\right\}\psi_{Lm}=0. \tag{26}$$

This equation is similar to the equation encountered in the solution of single-mode[4] and two-mode ring laser problems[5] except for the r-dependent factor multiplying the eigenvalue. Unfortunately Eq. (26) cannot be solved analytically and numerical methods have to be used. Finally, in terms of the new functions the two-time joint probability has the form

$$p_2(I_1,I_2,t+T|I_1',I_2',t)=[p_s(u)p_s(u')]^{1/2}\sum_{Lm}R_{Lm}^*(u)\left(1+\frac{B}{A}u\right)^{1/2}S_L^*(v)R_{Lm}(u')\times\left(1+\frac{B}{A}u'\right)^{1/2}S_L(v')e^{-\lambda_{Lm}T},\quad T\geq 0. \tag{27}$$

Higher-order joint probabilities can be expressed similarly. With the knowledge of the two-time joint probability function, we are now in a position to discuss the intensity correlations.

IV. INTENSITY CORRELATIONS

We shall consider only the second-order or the two-time intensity correlations. Higher-order correlations can be discussed similarly with the knowledge of the higher-order joint probabilities. From the definition and by symmetry, the two autocorrelation functions are equal, i.e.,

$$\langle I_1(t)I_1(t+T)\rangle=\langle I_2(t)I_2(t+T)\rangle=\int\int\int\int I_1'I_1\,p_2(I_1,I_2,t+T|I_1',I_2',t)dI_1dI_2dI_1'dI_2'. \tag{28a}$$

The intensity cross-correlation function is defined by

$$\langle I_2(t)I_2(t+T)\rangle=\langle I_2(t)I_1(t+T)\rangle=\int\int\int\int I_1'I_2\,p_2(I_1,I_2,t+T|I_1',I_2',t)dI_1dI_2dI_1'dI_2'. \tag{28b}$$

We can evaluate them all together by writing

$$\langle I_j(t)I_{j'}(t+T)\rangle=\int\int\int\int I_jI_{j'}'p_2(I_1,I_2,t+T|I_1',I_2',t)dI_1dI_2dI_1'dI_2',\quad j,j'=1,2. \tag{29}$$

With the help of Eq. (27) and Eqs. (18) and (19) we obtain

$$\langle i_j(t)I_{j'}(t+T)\rangle=\tfrac{1}{4}\sum_{Lm}\left|\int_0^\infty du\,u^2R_{00}\left(1+\frac{B}{A}u\right)R_{Lm}\right|^2\times\left(\frac{2l+1}{2}\right)^{1/2}e^{-\lambda_{Lm}T}\int_{-1}^1dv[1-(-1)^jv]P_l(v)\int_{-1}^1dv'[1-(-1)^{j'}v']p_l(v'),\quad j,j'=1,2. \tag{30}$$

From the orthogonality property of the Legendre polynomials, we have

$$\int_{-1}^1[1-(-1)^jv]P_l(v)dv=2\delta_{l0}-\tfrac{2}{3}(-1)^j\delta_{l1}. \tag{31}$$

Using Eqs. (25) and (31) in Eq. (30) we obtain the following expression for the intensity correlations:

$$\langle I_j(t)I_{j'}(t+T)\rangle = \sum_{m=0}\left[e^{-\lambda_{0m}T}\left|\int_0^\infty r^2 dr\, \psi_{00}\left(1+\frac{B}{A}r^2\right)\psi_{0m}\right|^2 + \frac{(-1)^{m+j'}}{3}e^{-\lambda_{2m}T}\left|\int_0^\infty r^2 dr\, \psi_{00}\left(1+\frac{B}{A}r^2\right)\psi_{2m}\right|^2\right], \quad j,j'=1,2. \tag{32}$$

The r integrals have to be evaluated numerically. The intensity correlations, thus, can be expressed as a series of falling exponentials with coefficients which can be evaluated in terms of the eigenfunctions of Eq. (27). The behavior of the normalized intensity correlation functions

$$\mu_{jj'}(T) = \frac{\langle I_j(t)I_{j'}(t+T)\rangle}{\langle I_j\rangle\langle I_{j'}\rangle} - 1, \quad j,j'=1,2 \tag{33}$$

is illustrated in Figs. 1 and 2. Numerical calculations show that near threshold many terms in the series of exponentials contribute and a single exponential approximation is rather poor. However, for large excitations they are well approximated by a single exponential. In fact,

$$\mu_{jj'}(T) \simeq \frac{(-1)^{j+j'}}{3}\exp\left(-\frac{2BC^2}{A(A-C)}T\right) \tag{34}$$

is a good approximation for $A \gtrsim 1.01C$. It follows from Eq. (34) that with increasing excitation intensity correlation time may increase which is clearly reflected in Figs. 1 and 2. These conclusions are similar to those reached by Tehrani and Mandel and Hioe[7] for a two-mode laser. This similarity again points out to the fact that they both correspond to a system of two neutrally coupled modes as noted in Ref. 1. In view of these remarks we expect that the general time-dependent problem when the losses of the two modes are not equal can be discussed using a perturbative approach for small differences in the losses along the lines of Ref. 5 and similar conclusions may be reached. We shall not consider this problem any further, however.

V. SUMMARY

We have considered and solved the problem of two-mode laser oscillations on coupled transitions in a Λ configuration of a three-level atomic system. Two-time intensity correlations have been studied and the expressions for them as a series

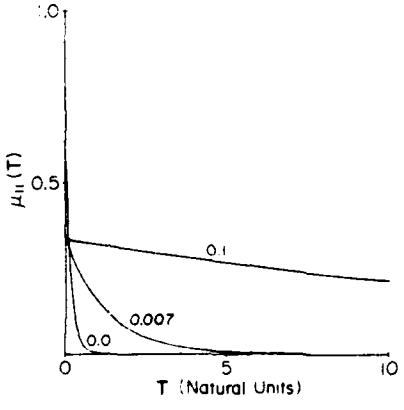

FIG. 1. Normalized autocorrelation function $\mu_{11}(t)$ as a function of T for three operating points $(A-C)/A = 0.0, 0.007, 0.1$. The parameters for this figure are $A = 1\mu s^{-1}$, $A/B = 10^6$ and C was varied.

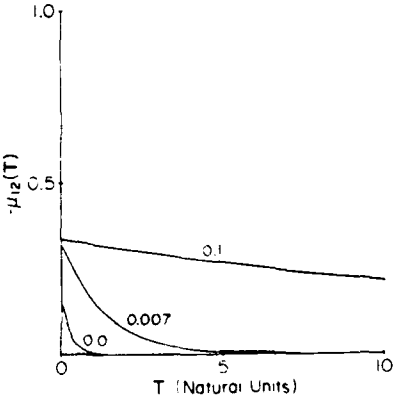

FIG. 2. Normalized cross-correlation function $\mu_{12}(t)$ as a function of T for three operating points. For parameters see Fig. 1.

of decreasing exponentials have been derived. It is found that the correlation time increases with increasing excitation and that for even moderately high excitations all second-order intensity correlations are well approximated by a single exponential.

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NONCLASSICAL EFFECTS IN A TWO-PHOTON LASER ☆

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The steady state quantum statistical properties of light in a two-mode two-photon laser are presented. It is shown that the second-order coherence functions violate a classical inequality.

In many systems involving the interaction between light and a medium, the quantum statistical properties of light are predicted to exhibit some nonclassical effects [1]. The most well-known example of these effects is photon antibunching which was predicted and then observed in resonance fluorescence [2,3] and theoretically predicted in a multiphoton absorption process [4,5], second-harmonic generation [6], degenerate parametric amplification [7], and free-electron laser [8], etc. We define the degree of second-order coherence of light to be

$$G_{ij}^{(2)} = \frac{\langle a_i^\dagger a_j^\dagger a_j a_i \rangle}{\langle a_i^\dagger a_i \rangle \langle a_j^\dagger a_j \rangle}, \tag{1}$$

where $a_i^\dagger$ and a_i are the creation and destruction operators of the field in the mode i and the angular brackets denote the ensemble average. The photon antibunching is exhibited by the fields which satisfy the inequality

$$G_{ii}^{(2)} < 1 , \tag{2}$$

i.e. the degree of second-order coherence is less than unity. This is true of the field distributions in which photons are anticorrelated.

In the case of intensity measurements on the two beams, we obtain [for a non negative two-mode Glauber's coherent state representation $P(u_1, v_2)$]

$$\iiiint (|u_1|^2|v_2|^2 - |v_1|^2|u_2|^2)^2 \times P(u_1, u_2)P(v_1, v_2)\, \mathrm{d}^2u_1\, \mathrm{d}^2u_2\, \mathrm{d}^2v_1\, \mathrm{d}^2v_2 \geq 0 .$$

This leads to the following quantum analogue of the Cauchy's inequality [1]

$$G_{12}^{(2)} < [G_{11}^{(2)} G_{22}^{(2)}] . \tag{3}$$

The violation of this inequality would be expected in systems where the correlation between the two beams is larger than the correlation between the photons of the same beam. This nonclassical correlation between the light beams has been observed in two-photon cascade emission [9].

In this paper we give another example where the violation of the inequality (3) could be observed.

We consider a two-mode two-photon laser in which lasing action is achieved by stimulated emission of two photons in a single atomic decay. The laser model consists of a coupled system of field and identical two-level atoms. The lasing levels of the atoms are assumed to have the same parity under the usual dipole approximation for the two-photon transition to take place.

The quantum statistical properties of the optical

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field of a two-photon laser have been studied in details in recent years [10–17]. These include studies on single-mode and two-mode lasers. Many authors restrict themselves to the model of a two-photon laser in which the end mirrors are assumed to be transmitting at the sum frequency [10,13,14,17]. The restriction to a two-photon loss mechanism is made in order to retain the property of detailed balance. In this paper we consider a more realistic model of a two-mode two-photon laser in which the cavity loss mechanisms of the two modes are simulated by the single photon processes.

We consider two field modes in a resonant cavity interacting with a system of identical two-level atoms. The atom–field interaction may be described by the effective hamiltonian in the interaction picture

$$H_I = \hbar g(\sigma^+ \hat{a}_1 \hat{a}_2 + \sigma^- \hat{a}_1^\dagger \hat{a}_2^\dagger) , \tag{4}$$

where g is the coupling constant and the atom operators σ^+ and σ^- are defined by

$$\sigma^+ = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}, \quad \sigma^- = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}. \tag{5}$$

The master equation for the photon distribution function $p(n_1, n_2)$ can be obtained by a straightforward extension of the Scully–Lamb theory [18] of a single photon laser. The resulting equation is

$$\frac{\mathrm{d}p(n_1, n_2)}{\mathrm{d}t} = \frac{-A(n_1+1)(n_2+1)}{1+(B/A)(n_1+1)(n_2+1)} p(n_1, n_2)$$

$$+ \frac{An_1 n_2}{1+(B/A)n_1 n_2} p(n_1-1, n_2-1)$$

$$+ C_1(n_1+1)p(n_1+1, n_2) - C_1 n_1 p(n_1, n_2)$$

$$+ C_2(n_2+1)p(n_1, n_2+1) - C_2 n_2 p(n_1, n_2) , \tag{6}$$

where A and B are the gain and saturation parameters for the two-photon gain mechanism and C_i ($i = 1,2$) is the loss parameter for the ith mode which is related to the mode frequency ω_i and Q of the cavity by the relation

$$C_i = \omega_i / Q . \tag{7}$$

This equation is difficult to solve even in steady state because each element $p(n_1, n_2)$ is coupled to $p(n_1 - 1, n_2 - 1)$, $p(n_1+1, n_2)$ and $p(n_1, n_2+1)$ which represent two-photon emission (one in each mode), single-photon absorption in mode 1 and single-photon absorption in mode 2 respectively.

We can however determine the second-order correlation functions in steady state when the laser is operating high above threshold. In this regime $n_1 n_2 B/A \gg 1$. When this condition is satisfied, we can neglect unity in comparison with the terms proportional to B/A in the denominators of the first two terms on the rhs of eq. (6). We then obtain (in steady state)

$$-(A^2/B)p(n_1, n_2) + (A^2/B)p(n_1 - 1, n_2 - 1)$$

$$+ C_1(n_1+1)p(n_1+1, n_2) - C_1 n_1 p(n_1, n_2) \tag{8}$$

$$+ C_2(n_2+1)p(n_1, n_2+1) - C_2 n_2 p(n_1, n_2) = 0 .$$

Various moments of n_1 and n_2, e.g. $\langle n_1^\alpha n_2^\beta \rangle$ ($\alpha, \beta = 0, 1, \ldots$) can be obtained by multiplying eq. (8) by $n_1^\alpha n_2^\beta$ and summing over n_1 and n_2. It can be easily shown that (for $i = 1, 2$)

$$\langle n_i \rangle = A^2/BC_i , \tag{9}$$

$$\langle n_i^2 \rangle = \langle n_i \rangle^2 + \langle n_i \rangle , \tag{10}$$

$$\langle n_1 n_2 \rangle = \langle n_1 \rangle \langle n_2 \rangle [1 + (\langle n_1 \rangle + \langle n_2 \rangle)^{-1}] . \tag{11}$$

On recalling the definitions of $G_{ij}^{(2)}$ [eq. (1)], we obtain

$$G_{11}^{(2)} = G_{22}^{(2)} = 1 , \quad G_{12}^{(2)} = 1 + [\langle n_1 \rangle + \langle n_2 \rangle]^{-1} . \tag{12}$$

It is evident that the Cauchy's inequality (3) is violated in the present case. This nonclassical effect is however very small, of the order of $(\langle n_1 \rangle + \langle n_2 \rangle)^{-1}$.

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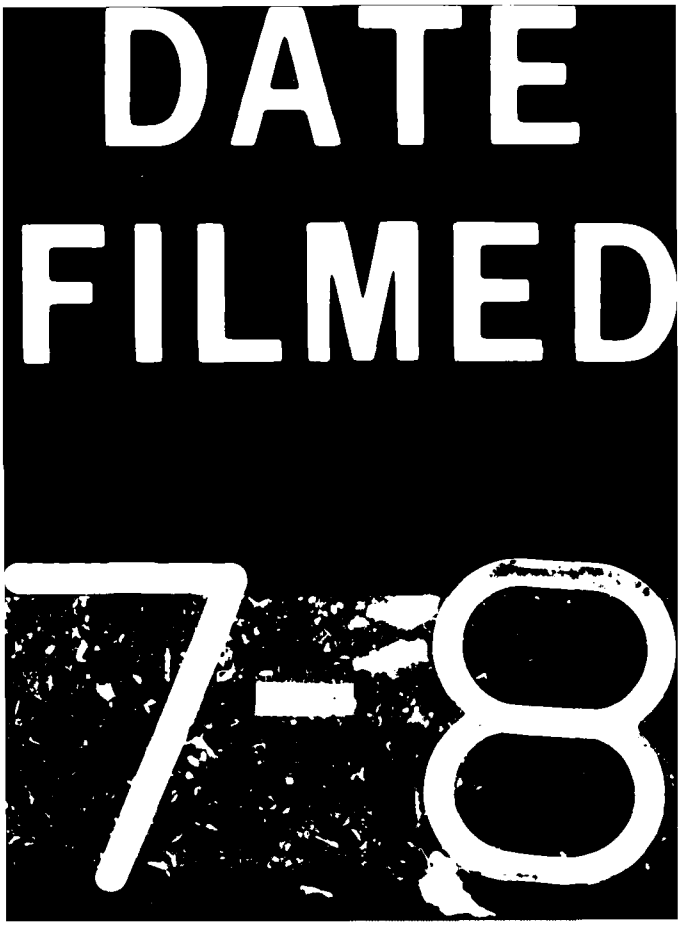
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